

Space Opportunities for Tropospheric Chemistry Research



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Space Opportunities for Tropospheric Chemistry Research

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PREFACE

The troposphere, the lowest region of the atmosphere, which extends from the surface to about 10 to 15 km (depending on latitude), contains about 85 percent of the total mass of the atmosphere. The troposphere is a region of great chemical diversity and activity, containing many dozens of trace gases produced by natural and anthropogenic processes at the surface and within the troposphere. Surface sources of trace gases include volcanism, biogenic production in soil and the ocean, biomass burning, internal combustion, and fossil fuel burning. Tropospheric sources of trace gases include atmospheric lightning, photochemical reactions initiated by solar photons, and atmospheric kinetic reactions. Trace gases in the troposphere impact the quality of the air that we breathe; lead to the production of acid precipitation; diffuse into the stratosphere, where they chemically destroy ozone; and control the climate of our planet via the "greenhouse effect." Over the last few years, we have found that several tropospheric trace gases--including methane, nitrous oxide, and carbon monoxide--which impact both atmospheric chemistry (in the troposphere and stratosphere) and climate appear to be increasing with time. Atmospheric environmental problems associated with tropospheric trace gases transgress national and international boundaries and are truly global in nature. The sensing, measuring, and monitoring of tropospheric trace gases from platforms in space offer a unique and important global vantage point. Over the next few decades, the planned Earth Observing System (EOS) and the Space Station will offer unique observing opportunities. Unfortunately, there are formidable technological and scientific problems associated with the measurement of tropospheric trace species from space.

A Workshop on Space Opportunities for Tropospheric Chemistry Research was held at the Goddard Institute for Space Studies in New York City, September 9 to 13, 1985, to address the technological and scientific problems associated with measuring tropospheric trace species from space. The objective of the workshop, sponsored by NASA's Tropospheric Chemistry Program, Earth Observation Division, Office of Space Science and Applications (OSSA), was to assess the near- and long-term technological and scientific problems associated with the measurement and monitoring of tropospheric trace gases and aerosols from space. The workshop considered different remote sensing techniques, including spectrometry, gas correlation filter radiometry, spectral radiometry, and lidar, as well as possible tropospheric missions. The workshop participants included atmospheric, measurement, and instrument scientists, technologists, engineers and mission planners from universities, industry, and Government laboratories. The workshop was structured to encourage free and open discussion. The first day of the workshop, a series of review papers were presented dealing with the scientific problems associated with tropospheric trace gases and aerosols, the state of the art of various remote sensing techniques, and space opportunities for measurement of tropospheric trace species. After the first day, the workshop participants were divided into instrument technology and mission panels. The ideas, conclusions, and recommendations of these panels were discussed before the entire workshop in plenary sessions and formed the basis of this report. The report was circulated in draft form to all workshop participants, and the comments received were incorporated into the final version, which was again circulated to all workshop chairmen for final review.

It is a pleasure to thank the panel chairmen, E. V. Browell, C. B. Farmer, R. S. Fraser, D. K. Killinger, D. G. Murcray, and H. G. Reichle, Jr., for their hard work and diligence in summarizing the panels' ideas, conclusions, and recommendations. I also thank the panel chairmen and F. Allario, D. Butler, R. J. McNeal, V. A. Mohnen, R. G. Prinn, and E. L. Tilton for preparing and presenting the overviews on the first day which set the stage for the entire workshop. Finally, it is a pleasure to

acknowledge the assistance and counsel of J. M. Hoell in all aspects of the planning of the workshop and the preparation of this report.

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SUMMARY

Tropospheric trace species (gases and aerosols) affect the photochemistry and chemistry of the entire atmosphere, as well as impact the climate of our planet. For numerous scientific and technological reasons, spaceborne measurements of tropospheric trace species have not kept pace with spaceborne measurements of trace gases in the mid and upper atmosphere. There are many formidable scientific and technological challenges to measuring tropospheric trace species from space. These challenges and possible solutions were the subjects of a Workshop on Space Opportunities for Tropospheric Chemistry Research which was held at the Goddard Institute for Space Studies in New York, September 9 to 13, 1985. This report summarizes the discussions, conclusions, and recommendations of this workshop. Workshop panels assessed the technology associated with spaceborne measurements of tropospheric trace species in the areas of spectrometry, gas filter correlation radiometry, spectral radiometry and tropospheric aerosols, and lidar technology and measurements.

Very recently, for the first time, a series of instruments demonstrated the feasibility of obtaining measurements of tropospheric trace species from space. Measurement of Air Pollution from Satellites (MAPS), a gas filter correlation radiometer in the nadir-viewing mode, obtained measurements of mid-tropospheric concentrations of carbon monoxide during two flights of the Space Shuttle--in 1981 and 1984. The Stratospheric Aerosol and Gas Experiment (SAGE II), an absorption spectrometer in the limb-viewing solar occultation mode launched on the Earth Radiation Budget Satellite (ERBS) in October 1984, obtained measurements of water vapor, ozone, and aerosols in the mid and upper troposphere. ATMOS (Atmospheric Molecules by Spectroscopy), a high-resolution interferometer in the limb-viewing solar occultation mode aboard SPACELAB-3 (April-May 1985), obtained measurements of several trace species down to the mid and upper troposphere. MAPS was the first dedicated space mission to obtain measurements of a tropospheric trace gas and demonstrated, for the first time, the tropospheric application of a gas filter correlation radiometer. The SAGE II and ATMOS measurements demonstrated, for the first time, that the limb-viewing solar occultation absorption technique has important applications to the measurement of trace species in the troposphere. Measurements of tropospheric aerosols are obtainable with several existing instruments, including the Advanced Very High Resolution Radiometer (AVHRR) on NOAA Polar-Orbiting Operational Environmental Satellites (POES), the Visible Infrared Spin Scan Radiometer (VISSR) on NOAA Geostationary Environmental Satellites (GOES), the multispectral sensors on Landsat, and SAGE II.

The workshop recommended that in the very near term, the proven MAPS, SAGE II, and ATMOS measurement techniques be exploited for measurements of tropospheric trace gases and species, including the analysis and interpretation of available tropospheric measurements. Also, in the near future, these proven instrument concepts should be expanded for possible additional capabilities of tropospheric measurements. In particular, feasibility studies should begin for the following instrument concepts: a three-layer (lower, middle, and upper troposphere) measurement of carbon monoxide and a lower or middle tropospheric measurement of methane using gas correlation radiometers in the nadir-viewing mode and a high-resolution interferometer operating in the nadir-viewing thermal emission model.

The workshop also considered the potential of spaceborne lidar systems for measurements of tropospheric trace gases and aerosols. Spaceborne lidar systems offer the potential of penetration into the lowest layers of the troposphere, as well as providing high vertical resolution of the distribution of tropospheric trace species. Spaceborne lidar systems for measurements of the vertical distributions of water

vapor, ozone, carbon monoxide, methane, ammonia, and aerosols, as well as column content measurements of tropospheric nitrogen dioxide, nitric oxide, and sulfur dioxide, were assessed. However, it was emphasized that the feasibility of such spaceborne lidar systems for tropospheric measurements has not been demonstrated to date, and considerable technology development will be required. Basic research and development that may lead to spaceborne lidar systems for tropospheric measurements include improved laser efficiency, lifetime, and spectral quality, as well as the development of the new lasers for tropospheric measurement applications.

INTRODUCTION: CHEMISTRY OF THE TROPOSPHERE

The troposphere, the lowest region of the atmosphere which extends from the surface to about 10-15 km (depending on latitude) and contains about 85 percent of the total mass of the atmosphere, is a region of great chemical activity. Tropospheric trace gases and aerosols control and/or impact:

- The quality of the air that we breathe
- Atmospheric visibility
- The formation of acidic precipitation
- The chemical destruction of ozone in the stratosphere
- The climate of our planet

Sources of tropospheric trace gases and aerosols include biogenic and anthropogenic activities, volcanoes, biomass burning, atmospheric lightning, and complicated atmospheric photochemical and chemical transformations. The sources and sinks and photochemistry and chemistry of tropospheric trace species have been discussed in a series of review papers (see, for example, Logan et al., 1981; Chameides and Davis, 1982; Levine and Allario, 1982; Levine, 1984; National Academy of Sciences, 1984; and Graedel, 1985). A summary of tropospheric trace species, concentration, atmospheric lifetime, and sources and sinks, as well as key tropospheric chemical and photochemical reactions grouped according to species family (i.e., oxygen, hydrogen, nitrogen, carbon, sulfur, and halogen species and aerosols), is given in appendix A.

It is interesting to note that over the last decade, almost all of the remote sensing of atmospheric trace gases from space involved the middle and upper atmosphere. The scientific and technological challenges of measuring tropospheric trace species from space are formidable and include:

- The presence of clouds and aerosols
- The presence of water vapor (H_2O) and carbon dioxide (CO_2), which blanket much of the useful infrared spectrum
- The very small temperature difference between the surface and the troposphere
- The high pressure of the troposphere which blends spectral features, making isolation of a particular signature very difficult

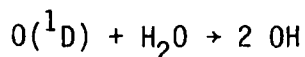
- The large abundance of certain stratospheric species--such as ozone (O_3) and nitrogen dioxide (NO_2)--which conceals tropospheric abundances of these species from space

A possible strategy for making measurements of tropospheric trace species from space was addressed in a recent National Academy of Sciences report, A Strategy for Earth Science From Space in the 1980's and 1990's, Part II. Atmosphere and Interactions With the Solid Earth, Oceans, and Biota (1985). This National Academy of Sciences report provided some recommendations as well as a list of the key tropospheric trace gases that should be considered for spaceborne measurement. (This list, which is reproduced in table I, provided an initial list of gases for measurement consideration by the workshop participants.) The National Academy of Sciences report recommended:

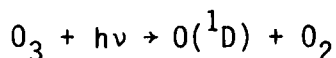
The primary objectives for the study of the troposphere from space are as follows, in order of priority:

1. To measure the magnitudes of the terrestrial and oceanic sources and sinks for radiatively and chemically important tropospheric trace gases, in particular, CO_2 , CO , CH_4 , and other hydrocarbons, N_2O , NH_3 , $(CH_3)_2S$, H_2S , OCS , and SO_2 .
2. To measure the atmospheric distributions, annual and latitudinal variations, and regionally and globally averaged long-term trends of chemically and radiatively important trace gases and aerosols, in particular, H_2O , CO , O_3 , NO_2 , SO_2 , and acidic or carbonaceous aerosols.

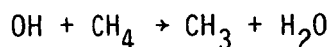
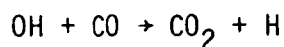
All four first-level tropospheric species (H_2O , O_3 , CO , and CH_4) in the National Academy of Sciences report are involved in the photochemical and chemical production and destruction of the hydroxyl radical (OH). The hydroxyl radical is the key chemically active species in the troposphere which reacts with almost every tropospheric species and, hence, controls the atmospheric lifetime of almost every tropospheric species. The only important tropospheric trace species whose lifetime is not controlled by reaction with OH is nitric oxide (NO), whose atmospheric lifetime is controlled by its reaction with O_3 . Although OH is the key chemically active species in the troposphere, its very short atmospheric lifetime (on the order of a second) and very low concentration (about 10^5 to 10^6 molecules cm^{-3}) have precluded reliable measurements with sufficient accuracy or precision for photochemical studies. Our knowledge about the concentration and distribution of OH is based on globally averaged measurements and a few in situ measurements in "non-remote" environments and on theoretical photochemical calculations. In the troposphere, the hydroxyl radical is formed by the reaction of excited atomic oxygen ($O(^1D)$) with water vapor (H_2O):



In the troposphere, excited atomic oxygen is produced by the photolysis of ozone (O_3):



The chemical destruction of OH is controlled by the reactions of OH with carbon monoxide (CO) and methane (CH₄):



Hence, we see that H₂O and O₃ lead to the photochemical production of OH, and CO and CH₄ lead to the chemical destruction of OH.

THE WORKSHOP: OBJECTIVES, AGENDA, AND PANELS

To consider the scientific and technological challenges and problems associated with the remote sensing of tropospheric trace gases and aerosols from space, the Tropospheric Chemistry Program, Earth Observation Division (EOD) of NASA's Office of Space Science and Applications (OSSA) held a workshop, "Space Opportunities for Tropospheric Chemistry Research," at the Goddard Institute for Space Studies, New York, September 9 to 13, 1985. (This workshop and report are the latest in a continuing series of NASA workshops and reports on tropospheric chemistry. The complete listing of the earlier workshops and reports is given in table II.) About 40 scientists--experts in atmospheric science, remote sensor instrumentation and technology, and mission planners--from universities, industry, and NASA participated in the workshop. A list of participants and their affiliations is given in appendix B.

The timing in the fall of 1985 for a workshop to consider and assess the measurement of trace tropospheric species from space was right for a number of reasons, including:

- In 1981, NASA sponsored a Conference on Tropospheric Passive Remote Sensing (see table II). Advances in passive remote sensing, as well as in active remote sensing (which was not addressed at all in the 1981 conference), have been significant over the last 5 years. The rapid scientific and technological progress in passive and active remote sensing over the last 5 years suggested that the time was appropriate for a workshop on this subject.
- The recent measurements by a series of satellite experiments (MAPS, SAGE II, and ATMOS) have demonstrated, for the first time, that certain tropospheric trace species can, indeed, be measured from space. (These satellite experiments are briefly summarized here.)

MAPS is a nadir-viewing gas filter correlation radiometer that flew on two flights of the Space Shuttle (November 1981 and October 1984) and obtained measurements of mid-tropospheric levels of carbon monoxide. MAPS, which is discussed in this report, was the first space mission dedicated to measurements of a tropospheric trace gas. Dr. Henry G. Reichle, Jr. of the NASA Langley Research Center is the Principal Investigator of MAPS (Reichle et al., 1982; and Reichle et al., 1986).

SAGE II is a limb-viewing absorption spectrometer in the solar occultation mode which was launched on the ERBS in October 1984. Although primarily

a stratospheric experiment, SAGE II obtained measurements of water vapor, ozone, and aerosols, down to the mid and upper troposphere. SAGE II is discussed in this report. Dr. M. Patrick McCormick of the NASA Langley Research Center is the Principal Investigator of SAGE II (Mauldin et al., 1985; and Woodbury and McCormick, 1986).

ATMOS, which is discussed in this report, is a Michelson interferometer in the solar occultation absorption mode that flew on SPACELAB 3 in April-May 1985. Like SAGE II, ATMOS is primarily a middle-to-upper atmospheric experiment that obtained measurements of a number of trace species down to the mid and upper troposphere. Dr. C. Barney Farmer of the Jet Propulsion Laboratory is the Principal Investigator of ATMOS (Farmer and Raper, 1986; and Park et al., 1986).

- The next several decades will be a period of rigorous planning and assessment by NASA of Earth observations from the Space Station and the dedicated Earth Observing System (EOS). EOS is a proposed Earth observation and information system, including new observing facilities and a data system to pursue a comprehensive multidisciplinary approach to studying the Earth as a system. EOS is tentatively scheduled for launch in the mid 1990's (NASA, 1984).
- New national and international concerns about increasing levels of tropospheric trace greenhouse gases that may impact the global climate of our planet developed (Kuhn, 1985; Ramanathan et al., 1985; Dickinson et al., 1986; and Wang et al., 1986).
- Recent measurements indicate that tropospheric levels of two chemically active species, methane and carbon monoxide, may have been increasing 1 to 2 percent over the last 35 years (Graedel and McRae, 1980; Rasmussen and Khalil, 1981; Khalil and Rasmussen, 1984; Rinsland et al., 1985; and Rinsland and Levine, 1985). This increase in methane and carbon monoxide may have caused a decrease in the tropospheric levels of the hydroxyl radical (OH) of as much as 25 percent over the last 35 years (Levine et al., 1985). As already noted, the hydroxyl radical is the key chemical scavenger in the troposphere which controls the atmospheric lifetime of almost every tropospheric species.
- An active ground-based/aircraft program of the chemistry and meteorology of the global troposphere, NASA's Global Tropospheric Experiment (GTE), was in full progress and providing new information on the composition and chemistry of the troposphere (McNeal et al., 1983; Journal of Geophysical Research, Special Section on the Global Tropospheric Experiment, 1985). The GTE is a key experiment to characterize different sources and sinks of tropospheric trace species, an important prerequisite to spaceborne measurements of tropospheric species.
- In 1984 and 1985, two relevant National Academy of Sciences reports appeared: (1) Global Tropospheric Chemistry: A Plan for Action (1984) and (2) A Strategy for Earth Science From Space in the 1980's and 1990's, Part II: Atmosphere and Interactions With the Solid Earth, Oceans, and Biota (1985).

The workshop was designed to consider, assess, and respond to these recent developments in tropospheric chemistry, advances in remote sensing technology, and future mission planning opportunities. All of the workshop participants met as a

group on the first day of the workshop and heard a series of 13 overview presentations covering the recent developments and activities in tropospheric chemistry, remote sensing instrumentation, and mission planning. These presentations included:

- Workshop Objectives: Dr. Robert J. McNeal, Manager, Tropospheric Chemistry Program, EOD, OSSA, NASA Headquarters
- Tropospheric Chemistry: An Overview: Dr. Joel S. Levine, Atmospheric Sciences Division, NASA Langley Research Center
- Tropospheric Chemistry: NASA and the National Program: Dr. Ronald G. Prinn, Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology
- The Space Station: Mr. E. Lee Tilton, Space Station Office, NASA Headquarters
- The Earth Observing System: Dr. Dixon Butler, EOD, OSSA, NASA Headquarters
- EOS and Tropospheric Chemistry: Dr. Volker A. Mohnen, Atmospheric Research Center, State University of New York at Albany
- Gas Filter Correlation Radiometry: Dr. Henry G. Reichle, Jr., Atmospheric Sciences Division, NASA Langley Research Center
- Interferometry: Dr. C. Barney Farmer, Earth and Space Science Division, Jet Propulsion Laboratory
- Spectrometry: Dr. David G. Murcray, Department of Physics, University of Denver
- Spectral Radiometry and Tropospheric Aerosols: Dr. Robert S. Fraser, Laboratory for Atmospheres, NASA Goddard Space Flight Center
- Lidar Measurements: Dr. Edward V. Browell, Atmospheric Sciences Division, NASA Langley Research Center
- Lidar Technology: Dr. Dennis K. Killinger, Lincoln Laboratory, Massachusetts Institute of Technology
- General Remote Sensor Technology Overview: Dr. Frank Allario, Flight Electronics Division, NASA Langley Research Center

On the second day of the workshop, all of the participants were assigned to participate in one of four instrument/technology panels. These four panels consisted of the following participants. (The affiliation and address of each participant are given in appendix B.)

- Spectrometry

C. B. Farmer and D. G. Murcray (Chairmen)
V. Abreu; J. C. Gille; R. A. Hanel
J. M. Hoell, Jr.; J. A. Jamieson; H. Zwick

- Gas Filter Correlation Radiometry

H. G. Reichle, Jr. (Chairman)
A. R. Barringer; R. Nichols; J. M. Russell III

- Spectral Radiometry and Tropospheric Aerosols

R. S. Fraser (Chairman)
M. Griggs; A. A. Lacis; L. R. McMaster

- Lidar Technology/Measurements

E. V. Browell and D. K. Killinger (Chairmen)
F. Allario; M. Bristow; H-Y Chiu; J. J. Degnan; D. E. Fitzjarrald
W. S. Heaps; C. L. Korb; J. B. Laudenslager, M. P. McCormick
I. S. McDermid; S. H. Melfi; V. A. Mohnen; R. Philbrick
A. Rosenberg; M. M. Sokoloski

The four measurement technology position papers form the bulk of this workshop report. As you will note, of the four measurement technology reports, the report from the lidar measurement technology panel is, by far, the longest. This is due to the fact that the 1981 NASA workshop only considered the role of passive remote sensors in measuring tropospheric trace species from space (see table I). Active remote sensors, i.e., lidar systems, were not considered at all in the earlier workshop.

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FUTURE INSTRUMENTATION AND MISSIONS FOR MEASUREMENTS OF TROPOSPHERIC TRACE
SPECIES FROM SPACE: WORKSHOP RECOMMENDATIONS

After the four instrument/technology panels completed their assessment of the available and projected instrument possibilities and the required technological/scientific activities to accomplish these objectives, all of the workshop participants were divided into three tropospheric mission panels which cut across instrument/technology lines. The mission panels were asked to: (1) recommend a phased space-measurement program to meet the scientific needs of an aggressive global tropospheric chemistry program for three timeframes: the present, the near term (1986-1992), and from 1992-2000--the period when the EOS is scheduled to become operational, and (2) recommend a supporting research and development program to ensure the successful accomplishment of the objectives of (1).

After their deliberations, the three mission panels reported their recommendations to the entire workshop. Although they differed in their approaches, the recommendations of the three mission panels were almost identical. Their recommendations and potential missions are summarized here and in table III.

The workshop missions panels recommended that gas filter correlation radiometry and high-resolution interferometry be exploited and expanded for measurements of the distribution of tropospheric trace gases. Such instruments and missions were identified and discussed by the panels and several instrument/measurement/mission feasibility studies were recommended by the panels, including:

- A three-layer (lower, middle, and upper troposphere) measurement of carbon monoxide, using a nadir-viewing gas filter correlation radiometer.

- A lower or middle tropospheric measurement of methane, using a nadir-viewing gas filter correlation radiometer.
- A survey of lower, middle, and upper tropospheric trace gases using a nadir-viewing high-resolution interferometer operating in the thermal emission mode.

The mission panels also concluded that lidar systems appear to have the potential of obtaining measurements of tropospheric trace gases and aerosols from space. In anticipation of the actual demonstration of spaceborne lidar measurements of tropospheric trace gases and aerosols, several lidar technology and measurement feasibility studies were considered, including:

- Feasibility studies to consider the potential of obtaining measurements of tropospheric trace gases and aerosols from spaceborne lidar systems. As discussed in this report, the Lidar Measurements and Technology Panel considered tropospheric measurements of the distribution of aerosols, water vapor, ozone, carbon monoxide, methane, and, possibly, ammonia, as well as column measurements of tropospheric nitrogen dioxide and, possibly, nitric oxide and sulfur dioxide.

In addition to the specific instrument/mission recommendations, the panels made several general recommendations that may lead to future spaceborne instruments, techniques, and missions for tropospheric chemistry research. This research may result in more accurate and/or faster algorithms and techniques for the retrieval and inversion of spaceborne radiative data for future tropospheric missions and in the possible development of new technologies for spaceborne measurements of tropospheric trace gases. The panels recommended research into the following areas:

- Basic studies in tropospheric radiative transfer and spectroscopy, e.g., line mixing in carbon dioxide, line shapes (especially for water vapor), and the behavior of the continuum.
- Basic research and development in detectors, coolers, and submillimeter and microwave components for the next generation of tropospheric spaceborne instrumentation; basic research and development aimed at improving existing laser efficiency, lifetime, and spectral quality and the development of new lasers for spaceborne measurements of tropospheric gases.

Spaceborne missions for tropospheric trace gases and aerosols presently available and projected for the future are summarized in table III.

A summary of the workshop, including a discussion of the scientific and technological challenges of obtaining measurements of tropospheric trace gases and aerosols from space, was reported in a paper presented at the American Meteorological Society Second Conference on Satellite Meteorology/Remote Sensing and Applications, Williamsburg, Virginia, May 13 to 16, 1986 (Levine et al., 1986).

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TABLE I.- TROPOSPHERIC TRACE GAS MEASUREMENTS FROM SATELLITES
(National Academy of Sciences (1985))

<u>Gas</u>	<u>Concentration (V/V)</u>
<u>First Level</u>	
H ₂ O	0.05- 0.04
O ₃	40 -100 ppb
CO	0.05- 0.3 ppm
CH ₄	1.7 ppm
<u>Second Level</u>	
N ₂ O	0.3 ppm
NO ₂	0.01- 10 ppb
NH ₃	0.1 - 1 ppb
SO ₂	0.05- 1 ppb
Chlorofluoromethanes	0.2 ppb
HC ₂	0.1 - 1 ppb

TABLE II.- NASA WORKSHOPS AND REPORTS ON TROPOSPHERIC CHEMISTRY
(Listed in Chronological Order)

Levine, J. S. and D. R. Schryer (eds.), Man's Impact on the Troposphere - Lectures in Tropospheric Chemistry, NASA Reference Publication 1022, 376 pages, 1978.

Seinfeld, J. H., F. Allario, W. R. Bandeen, W. L. Chameides, D. D. Davis, E. D. Hinkley, and R. W. Stewart, Report of the NASA Working Group on Tropospheric Program Planning, NASA Reference Publication 1062, 161 pages, 1981.

Schryer, D. R. (ed.), Heterogeneous Atmospheric Chemistry (Proceedings of a workshop held in Albany, NY, June 29-July 3, 1981), Geophysical Monograph 26, American Geophysical Union, Washington, DC, 274 pages, 1982.

NASA, Applying Modeling Results in Designing a Global Tropospheric Experiment (Proceedings of a workshop held in Virginia Beach, VA, July 15-16, 1981), NASA Conference Publication 2235, 94 pages, 1982.

Keafer, L. S. (ed.), Tropospheric Passive Remote Sensing (Proceedings of a workshop held in Virginia Beach, VA, July 20-23, 1981), NASA Conference Publication 2237, 94 pages, 1982.

NASA, Assessment of Techniques for Measuring Tropospheric N_xO_y (Proceedings of a workshop held in Palo Alto, CA, Aug. 16-20, 1982, 62 pages, 1983.

Hoell, J. M. (ed.), Assessment of Techniques for Measuring Tropospheric H_xO_y (Proceedings of a workshop held in Palo Alto, CA, Aug. 16-20, 1982), NASA Conference Publication 2332, 138 pages, 1984.

Crosley, D. R. and J. M. Hoell (eds.), Future Directions for H_xO_y Detection (Proceedings of a workshop held in Menlo Park, CA, Aug. 1985), NASA Conference Publication 2448, 65 pages, 1986.

TABLE III.- TROPOSPHERIC MISSIONS: 1985-2000

<u>Species</u>	<u>Instrument</u>	<u>Comments</u>
1. <u>Past and Present</u>		
CO (Mid Trop.)	MAPS	Flown on Space Shuttle-- two flights (Nov. 1981; Oct. 1984)
H ₂ O, O ₃ , Aerosols (Mid to Upper Trop.)	SAGE II	On ERBS (Launched Oct. 1984)
Several Gases (Mid to Upper Trop.)	ATMOS	SPACELAB 3 (April-May 1985)
Aerosols	AVHRR	NOAA Polar-Orbiting Opera- tional Environmental Satellite (POES)
	VISRR	NOAA Geostationary Operational Environ- mental Satellite (GOES)
	Multispectral Sensors	LANDSAT
2. <u>Potential Future Near-Term Missions (Through 1992)</u>		
CO (Three Layers: (Lower, Middle, and Upper Trop.)	Gas Filter Correla- tion Radiometer-- Nadir-Viewing Mode	Should be studied
CH ₄ (Lower or Middle Trop.)	Gas Filter Correla- tion Radiometer-- Nadir-Viewing Mode	Should be studied
Several Gases (Lower, Middle, and Upper Trop.)	High Resolution Inter- ferometer--Thermal Emission Mode)	Should be studied
Aerosols	LITE (Lidar in Space Technology Experi- ment	Planned for 1989 flight of Space Shuttle as a tech- nology demonstration

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V. Abreu; J. C. Gille; R. A. Hanel
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INTRODUCTION**

In 1981 a workshop was held in Virginia Beach, VA, to assess the role that passive remote sensing might play in tropospheric chemistry. Because the conclusions of that workshop are pertinent to this workshop, they will be summarized, and some data that have become available since that time will be presented. In figure 1 the possible passive remote sensing techniques are summarized. Note that the techniques are split into two viewing geometries--limb viewing and nadir viewing. Since the instrument requirements and capabilities are quite different for the two geometries, the instrument discussion was initially split according to the viewing geometry. It was further split as shown in the figure into radiometry and spectrometry. These categories were further subdivided according to instrument type.

The main advantage of limb viewing is the narrow weighting function (high-altitude resolution) and enhanced sensitivity due to the long atmospheric path. The major disadvantage is the lack of horizontal spatial resolution and, for tropospheric measurements, the probability that clouds in the field of view will limit the amount of data obtained. The geometry of the two paths and the probability of obtaining tropospheric data are shown in figures 2 and 3. Table IV shows the assessment made at the workshop of the potential of the various sensors for long-term measurements. The measurement requirements placed on the sensors at this workshop were fairly stringent as to the spatial coverage desired. Based on this requirement, limb viewing as a possible measurement technique was ruled out and emphasis was placed on nadir viewing. The various nadir-viewing techniques were compared. The results of this comparison are given in table V. Note that this table is based on the constraint of a narrow instrument field of view and an associated rapid detector response. The table gives the advantages and disadvantages of the various sensing techniques and also the technological thrusts needed for the instrument to meet the perceived measurement requirements.

In reviewing these conclusions, one finds that several things have occurred during the intervening period which significantly change some of the conclusions. The first is the realization that the solar occultation technique will yield data on the upper tropospheric distribution of several constituents of interest in tropospheric chemistry. The measurements obtained by the SAGE II satellite (particularly on upper tropospheric water vapor) show that the data are obtained on the tropospheric distribution often enough to give sufficient global coverage to be of interest in tropospheric chemistry. The recent results (Park et al., 1986) obtained by ATMOS, while a much smaller geographic data set, still demonstrated that data are obtained on the upper troposphere often enough to warrant use of such measurement techniques for tropospheric chemistry. When one agrees that limb-viewing geometry

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can be used for tropospheric measurement, then the instrument requirements change, particularly the spectral resolution which is required for the survey-type instrument. Recent results (Murcray et al., 1985) obtained with balloon-borne instruments also illustrate the point. Thus in figure 4, which shows several spectra obtained during sunset from balloon altitude, it is apparent that resolution of $\approx 0.02 \text{ cm}^{-1}$ is required to separate the spectral feature due to F-22 from the interfering absorption due to other compounds. Data concerning the spectral emission from the Earth's limb and also in the nadir-viewing mode have been obtained recently by two groups (University of Denver and Goddard Space Flight Center, Murcray et al., 1983; Murcray, 1984; Goldman et al., 1984; and Kunde et al., 1985). A portion of a limb scan obtained by the University of Denver group is shown in figure 5. This shows the strong emission due to F-12 in the lower stratosphere under this viewing geometry. Figure 6 shows this spectral region under nadir viewing. Note that although the feature is much less prominent, the absorption due to F-12 is clearly identifiable in the spectra. Again, resolution better than 0.1 cm^{-1} is needed to separate spectral features from interferences. These spectra also show the need for high-resolution spectral survey data to be obtained during early satellite missions in order to fully understand the data obtained by species specific instruments.

SOME SPECTROMETRIC TECHNIQUES[†]

Spectroscopic techniques are defined here as those methods which record a continuous portion of the emitted or transmitted spectrum containing spectral features of a number of tropospheric gases of interest; thus, the distinction is made between "species-specific" and "survey" instruments. While this classification may appear to be arbitrary from some points of view, its purpose becomes clearer in the context of the evolution of remote sensing techniques, in which a detailed knowledge of the absorption or emission spectrum is an essential precursor to the design of a species-specific instrument. The spectroscopic instruments of interest here include Grating spectrometers, Fourier Transform spectrometers (usually Michelson interferometers), and scanned Fabry-Perot interferometers.

Once the molecular constituents and the ancillary physical parameters to be measured have been clearly defined, together with their required spatial and temporal coverage, there is a strong motivation to develop methods which are optimized (in terms of the associated data rates) for these specific measurements. In general, spectroscopic techniques are not economic in their use of data transmission and storage systems when they are used to monitor single constituents or physical parameters. Their greatest value is in the early development phase of the program when the detailed spectral environment (i.e., local continuum characteristics, interference from other minor or trace constituents, etc.) must be understood in order that the design of a species-specific instrument and its data analysis procedures can be worked out correctly and with adequate understanding of the details of the radiative transfer in the chosen spectral region.

A question which arises in assessing the role of spectroscopic methods in remote sensing involves consideration of the modes of observation (i.e., limb versus nadir viewing, and emission versus absorption measurements) and the relative merits of the

[†]Prepared by C. B. Farmer.

available instrumental techniques (e.g., interferometers versus grating spectrometers). A complete discussion of the applicability of these options is beyond the scope of this document; as might be expected, however, each combination of instrument and observing technique has its own range of conditions over which it has superior performance. In general, absorption measurements can only be made in the limb-viewing, solar occultation mode and are thus limited to local sunrise and sunset times; against this they are relatively insensitive to the atmospheric temperature along the line of sight and are capable of high quantitative accuracy. Emission measurements can be made either viewing the atmosphere close to the limb to gain the sensitivity advantage of the long atmospheric paths given by this geometry, or down-looking (near-nadir viewing) to obtain better spatial resolution. Measurements made in emission are more prone to errors associated with uncertainties in the knowledge of the atmospheric temperature along the line of sight. Limb-viewing observations (in both emission and absorption) are capable of better vertical resolution (≈ 2 km) than are nadir observations (on the order of one pressure scale height); by contrast, the horizontal sampling dimension for limb observations is typically 200 to 300 km, whereas nadir observations can be made in some circumstances with horizontal scales on the order of 10 km.

Recently, several remote sensing spectroscopic techniques have been used successfully to fully record the composition of the troposphere from space, using principally limb-viewing methods (e.g., SAGE and ATMOS); the results have demonstrated the ability to make reliable quantitative determinations of the vertical distributions of both minor and trace constituents. Notable relevant examples include measurements of H_2O vapor, CH_4 , N_2O , halogen source species (CFMs, CCl_4 , etc.) and aerosols. The ATMOS experiment utilizes a Michelson interferometer in the limb absorption mode; figure 7 illustrates, in bar graph form, the large number of different molecular species seen in the ATMOS data and the altitude ranges over which their concentration profiles can be measured. Figure 8 shows an example of the data for a spectral region which includes an unresolved band of CFCI_3 (Freon 11).

While the usefulness of limb-viewing measurements is limited by the presence of tropospheric clouds, the experience gained thus far suggests that measurements in the upper troposphere (i.e., at altitudes above 5 km) are frequently possible, especially at mid-latitudes, and that all altitudes are accessible some of the time. Thus, the often-cited limitation of atmospheric opacity is far outweighed by the proven advantages of the limb-viewing absorption spectroscopy method, its inherent accuracy, good vertical resolution, and the wide range of species that can be measured.

In summary, spectroscopic measurements are required to define the spectral background and provide the detailed spectral information that is essential for the design of species-specific systems and the analysis of data obtained from them. This function of spectroscopic measurements is expected to be an important part of any tropospheric remote-sensing program, and both emission and absorption spectroscopy are relevant in this context. The data from such observations are of value to tropospheric science in their own right, during the initial phases while species-specific techniques and instruments are under development. In addition, there are a number of unresolved problems in tropospheric radiative transfer and spectroscopy (a few examples of which are line mixing in CO_2 , line shapes (especially for H_2O), and the behavior of continua) which presently limit the accuracy and reliability of all remote sensing methods. Only through a supporting program of spectroscopic measurements can progress be made in improving our understanding of these aspects of radiative transfer and ultimately reaching the desired confidence in the accuracy of species-specific monitoring techniques.

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TABLE IV.- PASSIVE REMOTE SENSING CONCEPTS

Generic sensor types	Potential long-term measurement roles		Heritage
<hr/>			
Species-specific sensors:	Long-term use of proven concepts for selected measurements:		
Optical filter radiometer radiometer	Nadir view:	Profiles of temp., H ₂ O vapor, Burden of ozone, aerosol optical thickness	SSH, HIRS TOMS, MSS, AVHRR SAM II, SAGE, LIMS
	Limb view:	Upper trop. profiles of aerosols and ozone	
Gas filter radiometer	Nadir view:	Two-layer measurements of CO, CH ₄	MAPS HALOE
	Limb view:	Upper trop. feasibility TBD	
Fabry-Perot filter radiometer	Nadir view:	Limited long-term role	CLAES
	Limb view:	Limited long-term role	
Correlation grating spectrometer	Nadir view:	Limited long-term role	COSPEC
Laser heterodyne techniques	Nadir view:	Geosync. orbit { Limited role in penetrating	IHR LHS
	Limb view:	Geosync. orbit { large strat. concentrations to trop. e.g., O ₃ , HNO ₃	
Spectral survey sensors:	Preferred long-term concepts for max. measurement capability:		
Grating spectrometer	Nadir view:	Many species and temp. profiles in 3.5- to 15-μm spectral range with spectral resolution of 0.1 cm ⁻¹ or better	SBUV SIRS B
	Limb view:		
Interferometer	Nadir view:		IRIS ATMOS
	Limb view:		

TABLE V.- COMPARISON OF NADIR-SENSING TECHNIQUES

Type	Advantages	Disadvantages	Technology needs
Gas filter radiometry			
All types	Low data rate Minimal data reduction High throughput (spectral and angular)	No detailed spectra Limited temperature profiles Test cells limited Species fixed after launch	Gas filter test cells Linear, high-dynamic-range detectors Highly uniform optical elements
Broadband spectrometry			
All types	Detailed spectral data Survey data Vertical discrimination	High data rate and processing Complex calibration Complex optical train Requires long stability Angle scan	On-board smart processing Cryogenics/cooling
Grating (with linear array)	Simple mechanism	Needs line array for sensitivity Large area gratings	10 ³ element arrays Large gratings
Interferometer	High throughput	Background fluctuations Lifetime of high-scan-rate reflector	Mitigation of background fluctuations Multiaperture, multi-band interferometer In-flight alignment verification
Narrow-band spectrometry			
Laser heterodyne spectroscopy	High spectral resolution for vertical discrimination	Limited tuning range	Tunable lasers and heterodyne arrays
Fabry-Perot	Simplicity	Limited tuning range Temperature stability	Improved coatings at long wavelengths

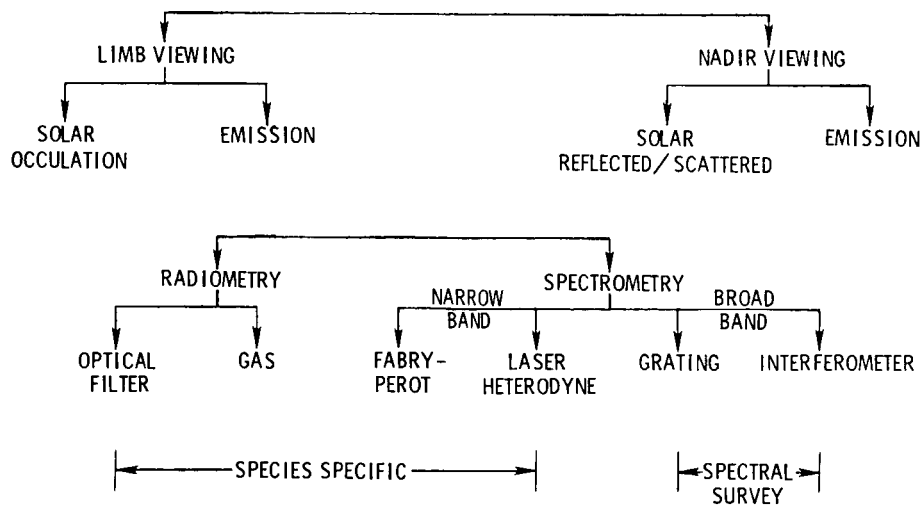


Figure 1.- Passive remote sensing tree showing generic techniques, viewing geometries, and associated sources.

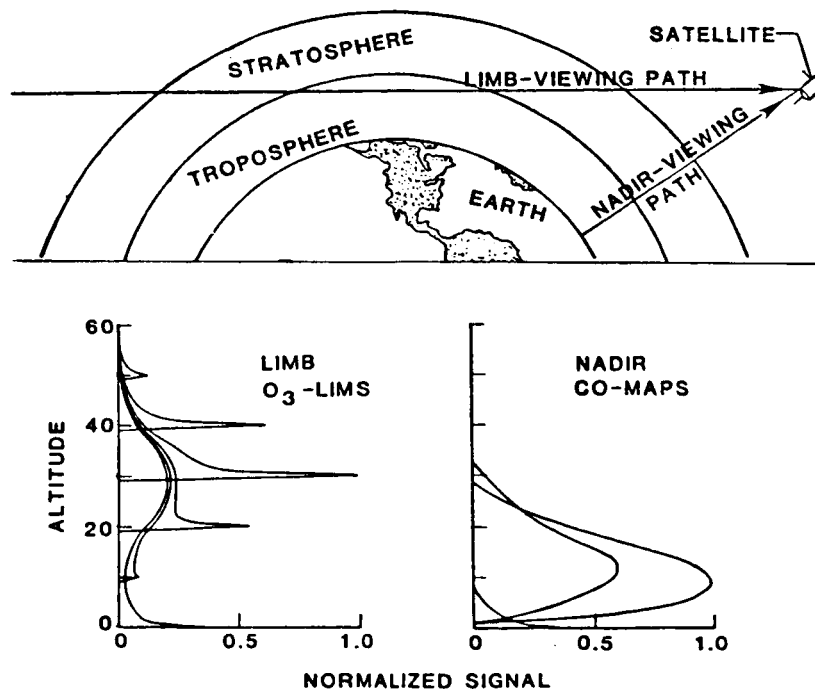


Figure 2.- Comparison of limb and nadir viewing.

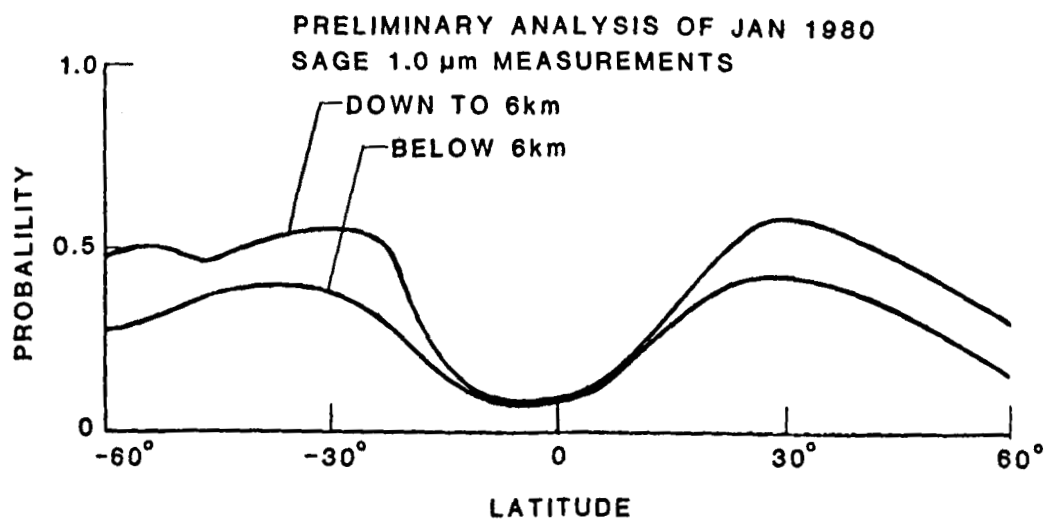


Figure 3.- Probability of limb-viewing measurements in the troposphere.

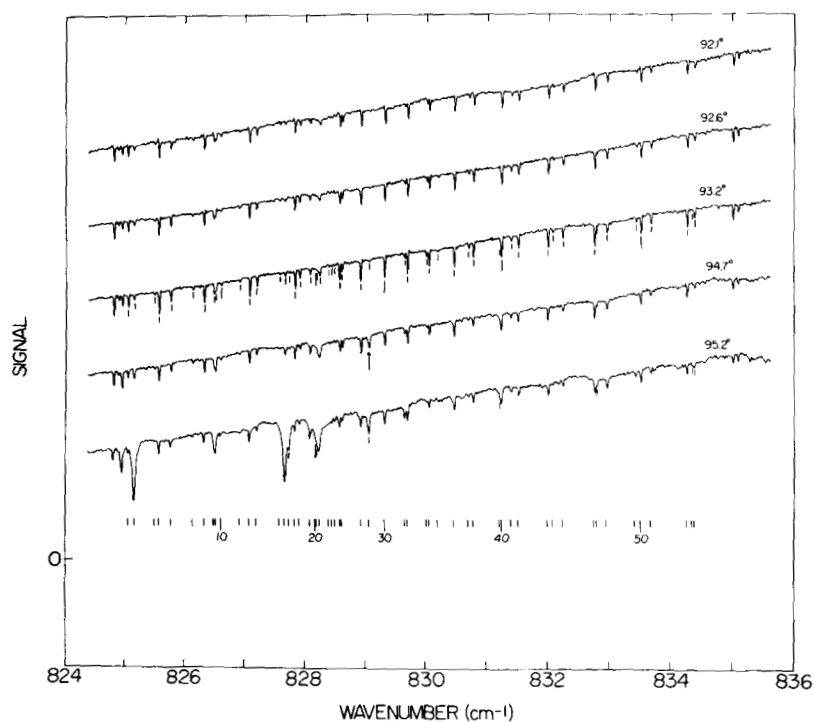


Figure 4.- Solar spectra in the $825\text{--}835\text{ cm}^{-1}$ region, obtained during a balloon flight on 23 March 1981, from Holloman AFB, New Mexico, with the University of Denver interferometer system at $\approx 0.02\text{ cm}^{-1}$ resolution. The altitude and solar zenith angles are indicated on the spectra. Zero levels are offset for clarity. The 829 cm^{-1} F-22 Q-branch is marked with an arrow.

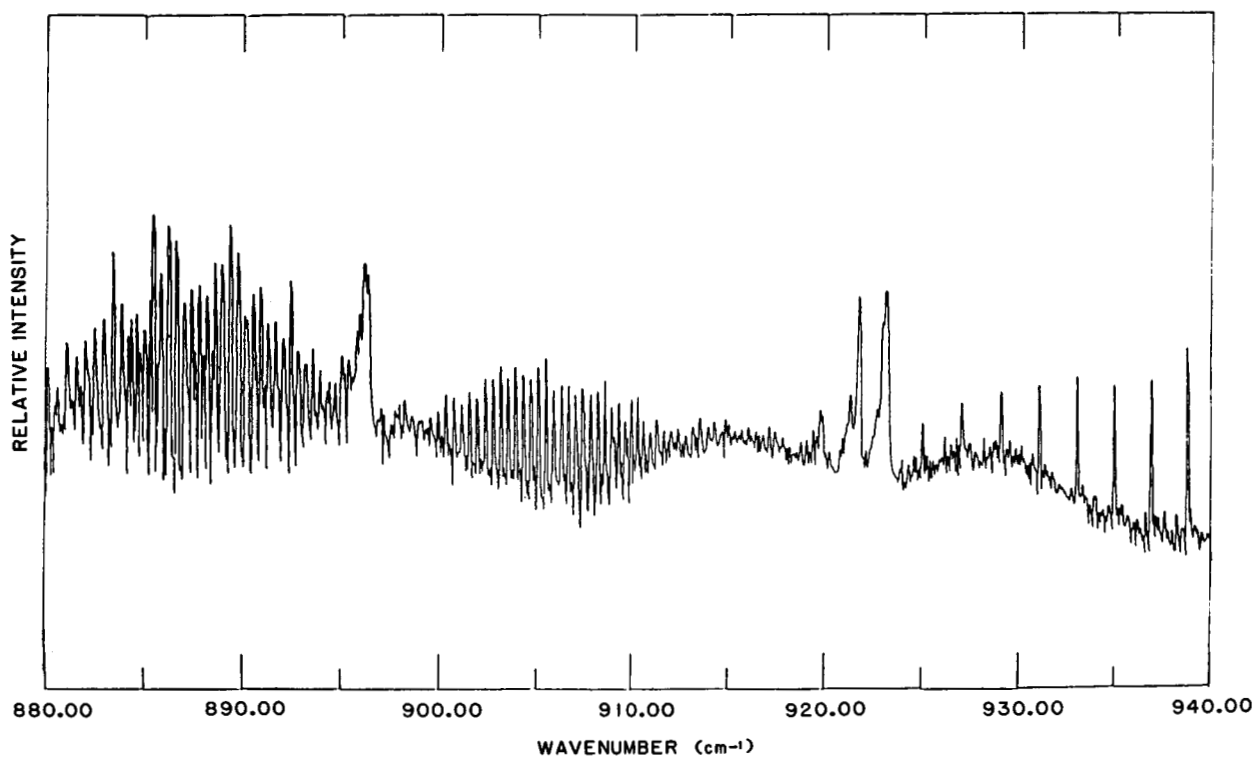


Figure 5.- Atmospheric emission from the Earth's limb as observed from a balloon. Balloon altitude 30 km, viewing angle $-3\frac{1}{2}^{\circ}$.

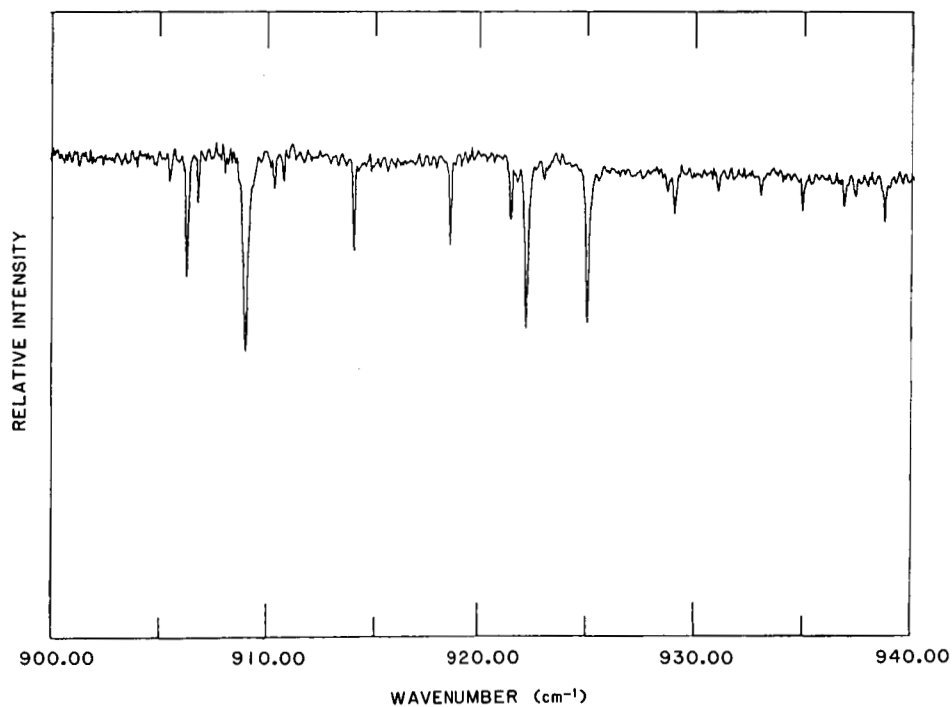


Figure 6.- Earth emission spectra as observed from a balloon at 30 km.

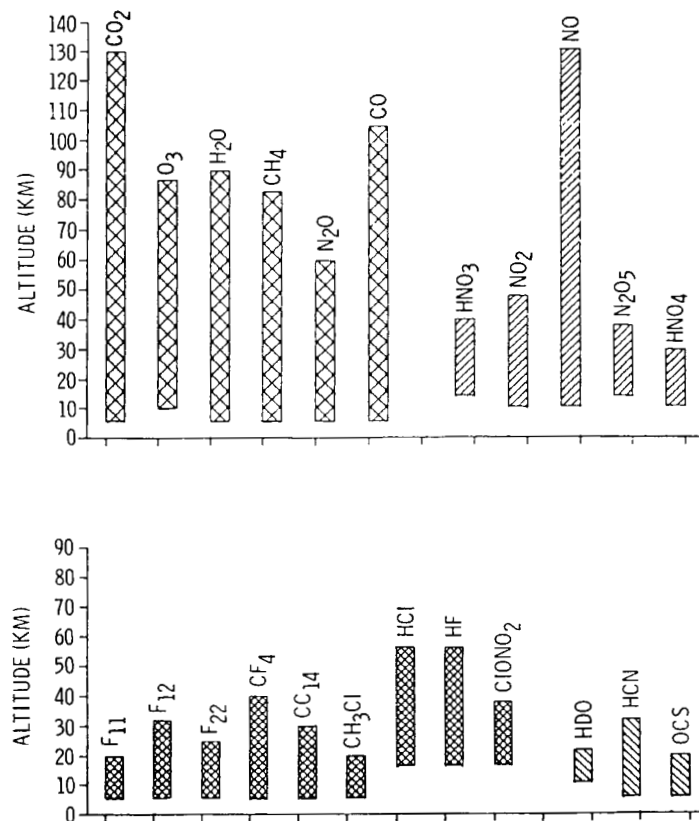


Figure 7.- Atmospheric minor and trace species derived from the ATMOS data, separated into chemical families and indicating the altitude regions over which profiles of concentration have been retrieved.

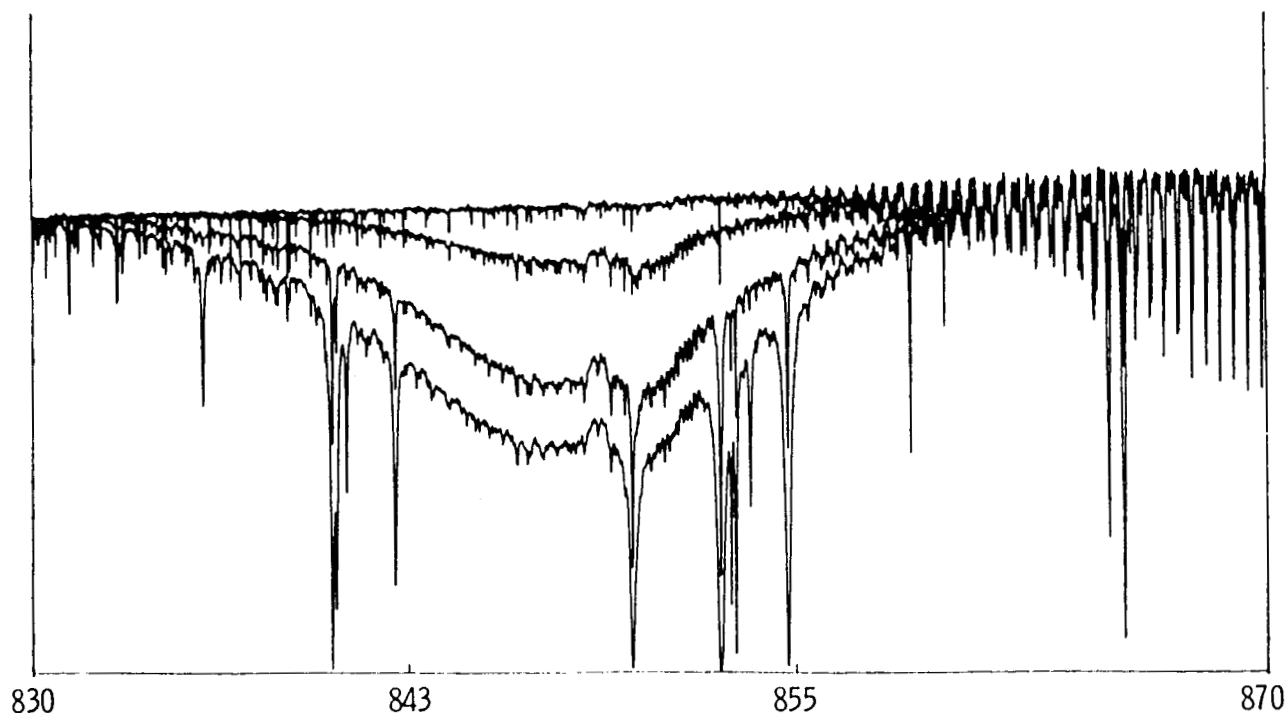


Figure 8.- Portions of four spectra in the region of 830 to 870 cm⁻¹ showing large unresolved spectral feature of Freon 11 in the lower three traces which were made at tangent altitudes of 15, 9, and 5 km.

GAS FILTER CORRELATION RADIOMETRY: REPORT OF PANEL

H. G. Reichle, Jr.* (Chairman)

A. A. Barringer, R. Nichols, and J. M. Russell III

To measure the concentration of a gas in the troposphere, the gas filter radiometer correlates the pattern of the spectral lines of a sample of gas contained within the instrument with the pattern of the spectral lines in the upwelling radiation. It has very-high-energy throughput, and it is able to take advantage of multiplexing since all wavelengths within the rather broad passband reach the detector at the same time. The effective resolution can be made very high since it is determined by the properties of the internal gas cell. Because the instrument does use internal gas cells as sources of the reference spectra, any single instrument can only measure a limited number of gases. In general, instruments can be built that are rugged and lightweight, require modest amounts of power, and have low data rates.

A schematic diagram of a generalized gas filter radiometer is shown in figure 9. Energy that has been emitted by or reflected from a background target is transmitted through the atmosphere to the instrument. The individual gases within the atmosphere selectively absorb and re-emit the energy in a pattern of lines that is unique to each particular gas. If the atmosphere is cooler than the radiation source, which may be either the Sun or the surface of the Earth, the gas removes a net amount of energy from the beam, and the lines are seen as absorption lines. If the atmosphere is warmer than the source, the gas contributes a net amount of energy to the beam, and the lines are seen as emission lines. If the gas in the atmosphere is at the same temperature as the radiation source, the gas emits an amount of energy equal to that which it absorbs and, hence, the atmospheric gas cannot be seen. Clearly, experiments that use reflected sunlight always operate in absorption. After being gathered by the foreoptics, the energy passes through the gas cell modulator to the detectors. In most of the instruments now being used, the signal is synchronously detected at the modulator frequency at the detector output. This signal is related to the degree of correlation between the absorption lines in the incoming energy and the absorption lines of the gas contained in the modulator.

The instruments currently in use can be divided into two basic types, depending upon the type of gas cell modulator used. Instruments of the first type are generally based upon the principle used in the Selective Chopper Radiometer (SCR) first flown on the Nimbus 4 satellite as a stratospheric temperature sounder (Abel et al., 1970). In this implementation of the principle, the incoming radiation is chopped between a fixed pressure and path-length cell containing a sample of the gas of interest and a vacuum cell or another gas cell containing a sample of the gas at a pressure different from that of the first cell. The instrument can be arranged so that the incoming energy passes alternately through the two cells to a single detector or so that the beam is divided and passes through the two cells simultaneously to two separate detectors. Both arrangements have been used to measure carbon monoxide in the atmosphere from an aircraft platform (Acton et al., 1973; and Reichle et al., 1986a). Only the second implementation has been used to measure tropospheric carbon monoxide from a satellite platform (Reichle et al., 1986b). The major advantages of

*Prepared panel report.

the two-cell modulator are simplicity and freedom that one has in selecting cell lengths, pressures, and materials. The major disadvantage of the two-cell modulator is the difficulty in insuring that the two optical legs of the instrument are maintained in a balanced condition; that is, for example, that their optical characteristics do not change differently as a result of environmental influences. Such differential changes usually result in zero-shift related errors. In order to circumvent the differential changes that occurred in early two-cell instruments, a single optical-path instrument was developed at Oxford University. Called the pressure-modulated radiometer (or p.m.r.), this device contains only a single cell, and it is the second type modulator in use. The transmission characteristics of the gas in the cell are changed, or modulated, by means of a piston arrangement that sinusoidally changes the cell pressure. This device has been applied to the measurement of stratospheric and mesospheric temperature and composition in a number of experiments flown aboard the Nimbus satellite series by the Oxford group. The latest of these was the Stratospheric and Mesospheric Sounder (SAMS) experiment flown aboard Nimbus 7. Because of its single optical path, this modulator is inherently free from the difficulties encountered in attempting to balance two different optical paths. Its major disadvantage, in its present implementation at least, is the limited range of mean pressures and pressure differences that can be achieved. A version of this device was evaluated as a candidate for application to the remote sensing of tropospheric species. The results of that study are summarized in Orr and Rarig (1981). The overall conclusions of the study were that, because of the limitations of mean pressure and pressure ratio, the application of the p.m.r. to tropospheric remote sensing was limited.

The next section describes three instruments that have application to remotely measuring tropospheric constituents.

CURRENT GAS FILTER RADIOMETERS

The Gas Filter Radiometer (GFR)

The MAPS experiment flown on the second flight of the Space Shuttle during November 1981 measured the tropospheric carbon monoxide mixing ratio along the sub-satellite track over a period of 12 hours between 38°N latitude and 38°S latitude. Since this was the first measurement of this gas made from a space platform, the instrument will be described in somewhat greater detail than will the other previously mentioned instruments.

The instrument layout is shown in figure 10. This instrument is of the SCR type in that it differences the signal as seen through two fixed cells. The energy enters the instrument through the objective lens and is chopped against a reference blackbody by a rotating chopper. After passing through a field stop, a beam combiner (the function of which will be discussed later) an aperture stop and a broad-band filter. The beam is divided by a series of beamsplitters, and it passes to three detectors. Before reaching two of the detectors, the energy passes through 1-cm-long gas cells. One of these is filled with 76 Torr of pure CO, while the second is filled with 266 Torr of pure CO. The outputs of the three detectors are differenced to form two difference signals, $\Delta V = V_{D3} - V_{D2}$ and $\Delta V' = V_{D1} - V_{D2}$. The output of detector D2 is also demodulated to form a broad-band radiometric signal called V that is indicative of the temperature of the underlying surface. Two internal blackbodies called balance sources are chopped at twice the signal frequency by the rotary chopper. The radiometric difference signal from these targets is combined via the previously mentioned beam combiner and directed onto the detectors. After demodulation at the

detectors, this signal is used to drive an automatic gain control circuit that maintains the instrument in the balanced condition. In spite of the extreme temperature variations encountered on the Space Shuttle flight (caused by the failure of a coolant loop temperature controller), this instrument performed very well.

Because the instrument views the nadir in the $4.67\ \mu\text{m}$ band, the signal received by the instrument is integrated over the depth of the atmosphere. The variation of the signal received, as a function of altitude, is described by a signal function, which is analogous to the weighting function as used in describing the performance of radiometric temperature sounders. The signal functions for the two channels as flown on the STS-2 are shown in figure 11. Since the signal received is proportional to the integral of these curves, it can be seen that both channels receive the majority of their signal from the middle and upper troposphere. Neither channel receives much information from the mixing layer because of the low thermal contrast between the atmosphere and the surface of the Earth.

Figure 12 shows a typical orbit of CO data produced by the MAPS instrument. (Because the sampling rate is high at $1\ \text{sec}^{-1}$, the individual data points are not resolved on this figure.) The open spaces in the trace are caused by the presence of clouds in the field of view, which renders the data nonreducible.

To better demonstrate their global-scale patterns, the data were averaged over 5° latitude by 5° longitude grid squares. The data from the higher pressure channel presented in this way are shown in figure 13. The strong variations of the mixing ratio with both latitude and longitude in the Tropics and in the Northern Hemisphere are clearly shown. These global-scale patterns in the CO data, which are so evident in the satellite data, would have been very difficult to measure, either from surface-based or aircraft-based platforms.

The Halogen Occultation Experiment (HALOE)

Another gas filter radiometer similar in principle to the MAPS instrument has been built by NASA as part of the Upper Atmosphere Research Satellite (UARS) which will be launched during 1989. This instrument measures vertical profiles of several constituents in the stratosphere by viewing the Sun during sunrise and sunset as seen from the spacecraft. A schematic diagram of the instrument is shown in figure 14. Because the Sun is now the radiation source, the field of view of the instrument is much narrower. Only a single balance source is required. As can be seen from table VI, the instrument has eight channels--four are gas-filter channels and four are broadband radiometer channels, and these channels span the range from 2.5 to $10\ \mu\text{m}$. A dichroic beamsplitter is used to separate the radiation prior to its passage through the gas cells. At the present time, this instrument represents the highest development of an SCR-type gas filter radiometer as a spaceborne remote sensor.

Gas Filter Correlation Spectrometer Developed at Centre for Research for Experiment Space Science (GASCOFIL)

While this instrument is not now being developed for a space application (as are MAPS and HALOE), it incorporates certain new ideas (or advancements on old ideas) that cause it to be important for any new instrument development. The instrument, like the original SCR, is a single-detector instrument rather than a multiple-detector instrument as are MAPS and HALOE. In this implementation of the technique, the difference signal is formed on the single detector by varying the cell that is in

the optical path. This was previously done by means of a rotary or vane chopper and a system of mirrors. In the GASCOFIL instrument, this is accomplished by mounting the gas cells and vacuum cells in a rotary wheel and rapidly spinning the wheel through the optical path. The detector then sequentially "sees" the scene through the gas cells and vacuum cells. Between each gas cell and/or vacuum cell view of the scene, the detectors "see" the spokes of the wheel. The output of the detector is amplified and immediately digitized. The signal corresponding to each cell or spoke is then stored in a separate memory location of a microprocessor-based computer. Because all signals are handled as digital rather than analog data, the problems of amplifier drift are greatly reduced. Further, it is possible to adjust certain parameters (averaging time, for example) in the software of the microprocessor rather than the hardware. This considerably increases the flexibility and adaptability of the instrument.

The GASCOFIL instrument has been operated in a portable mode to measure SO_2 in a power plant plume in Ontario, Canada, with excellent results. This application used wavelengths in the near UV. Tests to measure CO using the $4.67 \mu\text{m}$ band are planned.

CAPABILITY FOR REMOTELY SOUNDING TROPOSPHERIC CARBON MONOXIDE

A set of preliminary calculations to determine the feasibility of performing a multiple-layer, tropospheric carbon monoxide measurement experiment has been carried out. The calculations assume a U.S. Standard Atmosphere for the temperature profile and a typical value for the CO mixing ratio. The air is assumed to be in thermal equilibrium with the underlying surface, which was either land or water. Both the 4.67 and $2.33 \mu\text{m}$ bands were investigated. The line-by-line radiative transfer program was the same as that used in the reduction of the MAPS data. The results of these calculations are shown in figure 15 and table VII. It can be seen that a three-layer measurement in the troposphere is feasible. The signal functions for the two higher altitude channels are generally similar to those of the OSTA-1 MAPS experiment; they have been made somewhat narrower by varying the cell conditions. The signal levels are of the same order as those achieved by the MAPS experiment. To make measurements in the mixing layer where the temperature contrast between the surface and the gas is small, it is necessary to use reflected sunlight in the $2.33 \mu\text{m}$ region. (This, of course, limits the measurements to daytime only.) The signal levels are now about 2 orders of magnitude lower than those obtained in the longer wavelength channels. Measurement of the reflected sunlight component will require some engineering advance over the MAPS instrument, both in terms of noise level and stability. This will require some engineering development, but it should require no new technology. The calculations summarized in figure 15 and table VII assume a generic gas filter radiometer. Therefore, any instrument capable of chopping between the indicated cell lengths and pressures with the appropriate noise level and stability would be capable of making the measurement.

RECOMMENDATIONS

This group feels that the following is an appropriate evolution of a tropospheric measurement program using gas filter radiometry.

Now to 1992

1. Exploit HALOE and/or GASCOFIL technology to design, build, and fly a three-layer tropospheric CO measurement and to fly it on a NOAA operational satellite.
2. Test the feasibility of measuring CH₄ by modifying the current MAPS instrument and flying on a Space Shuttle flight.
3. Carry out studies to determine the feasibility of measuring other trace gases.

1992 - 2001

1. Build a multiple-level CO instrument for EOS that includes the capability of measuring CH₄ and other gases as shown feasible via numerical simulations and flight tests.
2. Perform feasibility studies of an instrument that replaces the gas cell with an etalon; O₃ and NH₃ are candidate gases.

2001 - Beyond

1. Build advanced instrument based on results of number 2 above.

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TABLE VI.- HALOE INSTRUMENT CHARACTERISTICS

GAS	CENTER (CM^{-1})	50% POINTS (CM^{-1})		MAX 5% POINT WIDTH
HF	4078	4047 \pm 6	4109 \pm 6	117
HC ₂	2940	2910 \pm 6	2970 \pm 6	108
CH ₄	2891	2870 \pm 6	2912 \pm 6	82
NO	1900	1883 \pm 6	1917 \pm 6	62
CO ₂	3572	3537 \pm 6	3608 \pm 6	128
H ₂ O	1514	1506 \pm 6	1522 \pm 6	29
O ₃	996	976 \pm 6	1017 \pm 6	78
NO ₂	1600	1591 \pm 5	1607 \pm 5	29

TABLE VII.- SUMMARY OF SIGNAL CHARACTERISTICS (ADVANCED SOUNDER)

GAS	ALTITUDE OF SIGNAL PEAK	BAND	SIGNAL LEVEL
CO	0 mb	2.3 μm	$2 \times 10^{-9} \text{ w/cm}^2 \text{ sr}$
CO	450 mb	4.67 μm	$2.5 \times 10^{-7} \text{ w/cm}^2 \text{ sr}$
CO	200 mb	4.67 μm	$1 \times 10^{-7} \text{ w/cm}^2 \text{ sr}$

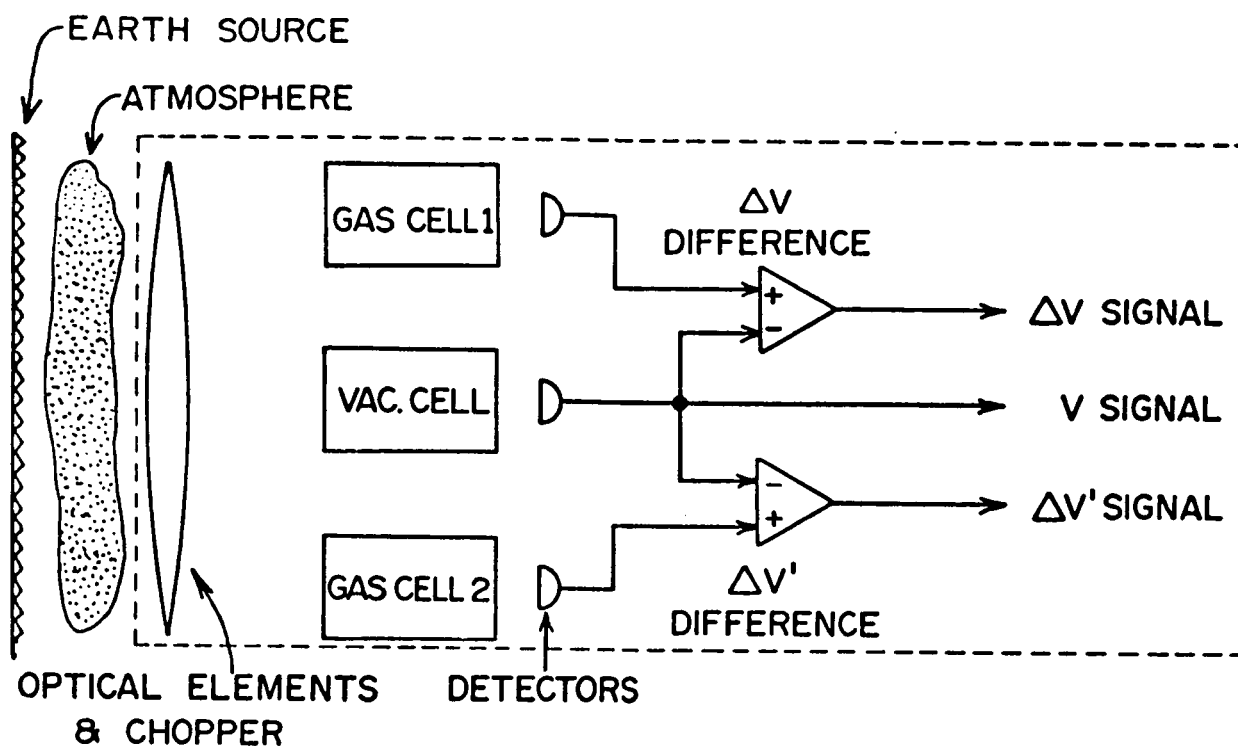


Figure 9.- GFR schematic.

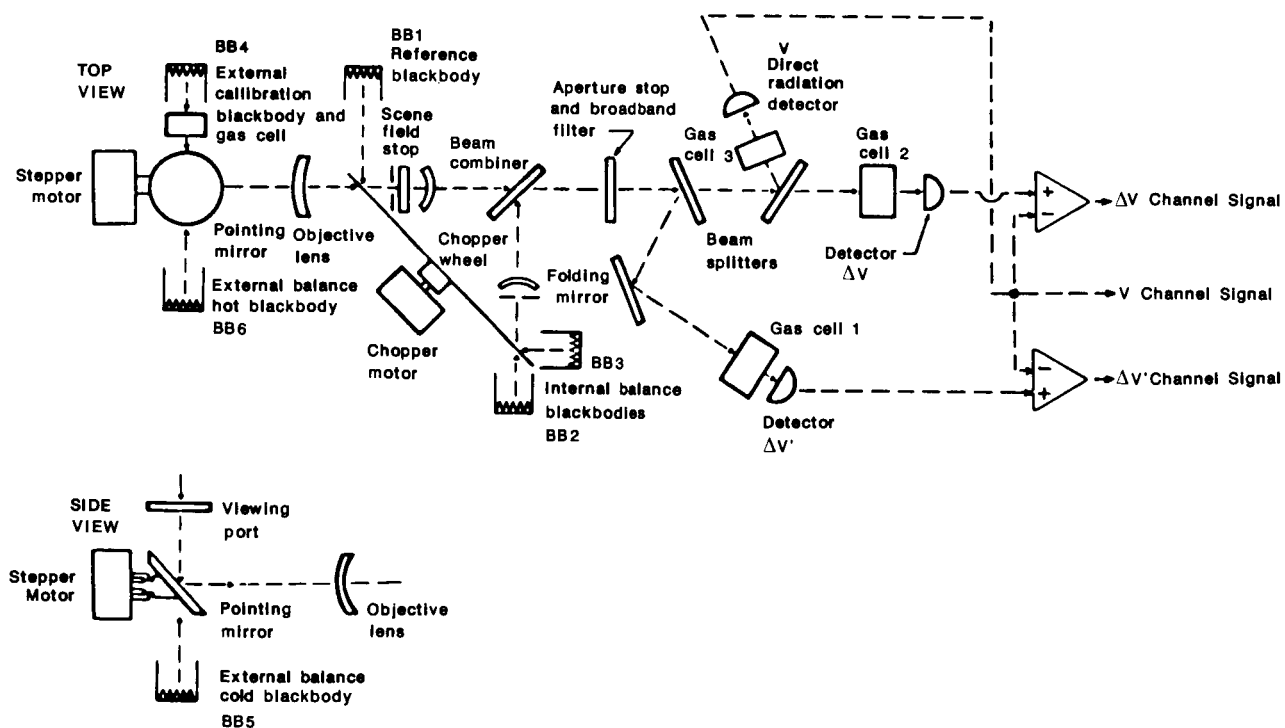


Figure 10.- MAPS instrument layout.

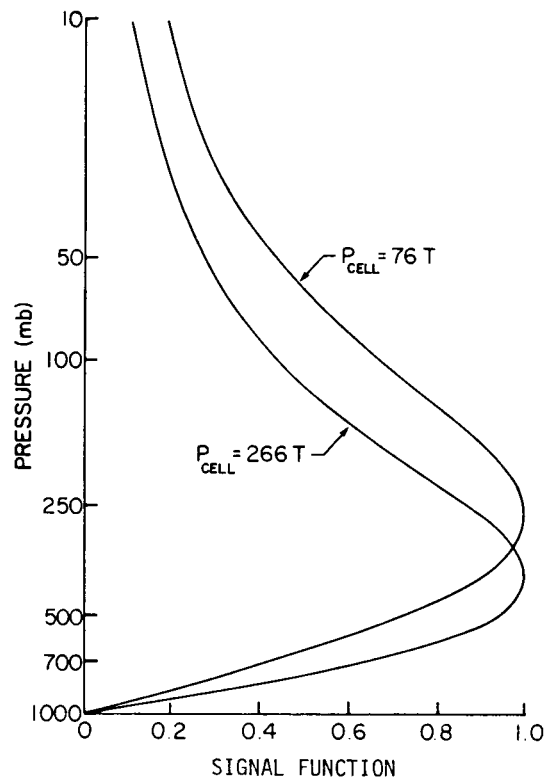


Figure 11.- MAPS OSTA-1 signal functions.

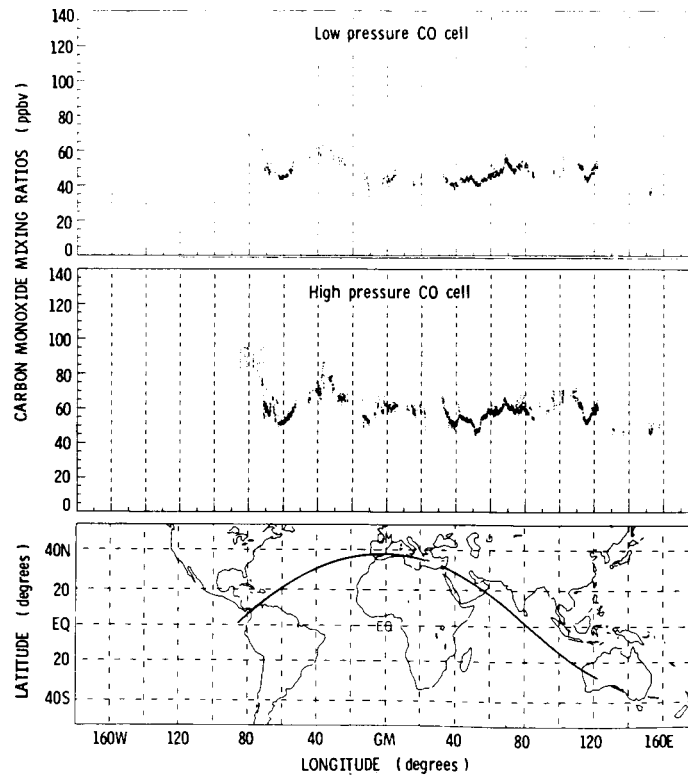


Figure 12.- MAPS data (1 orbit).

MAPS OSTA-1 CO(ppbv) NOV. 14, 1981

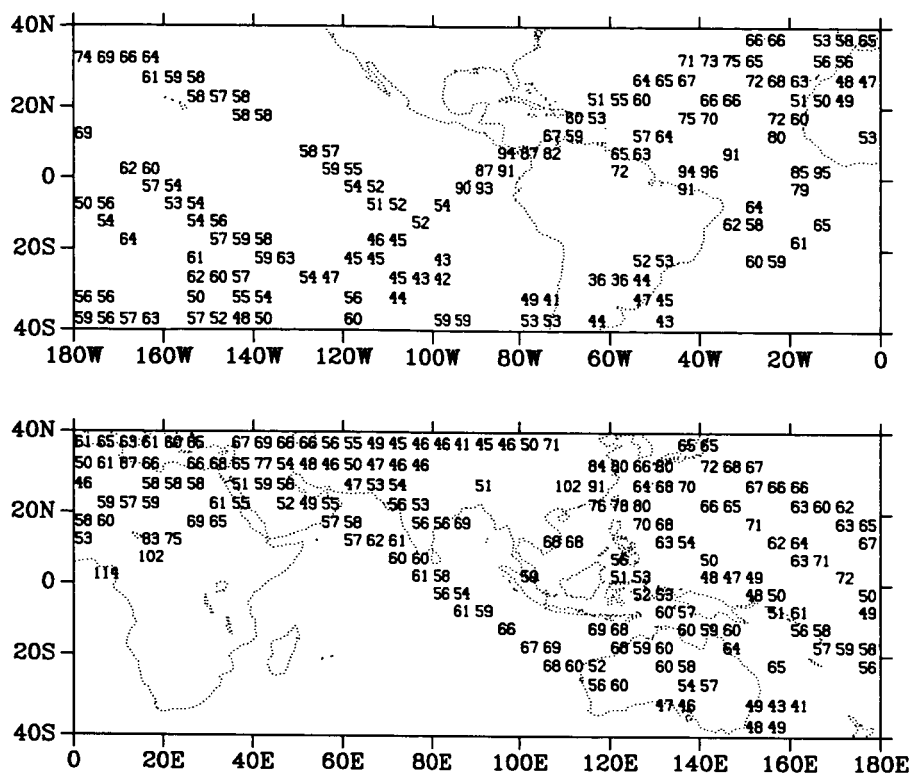


Figure 13.- MAPS data (5° x 5°) 266T.

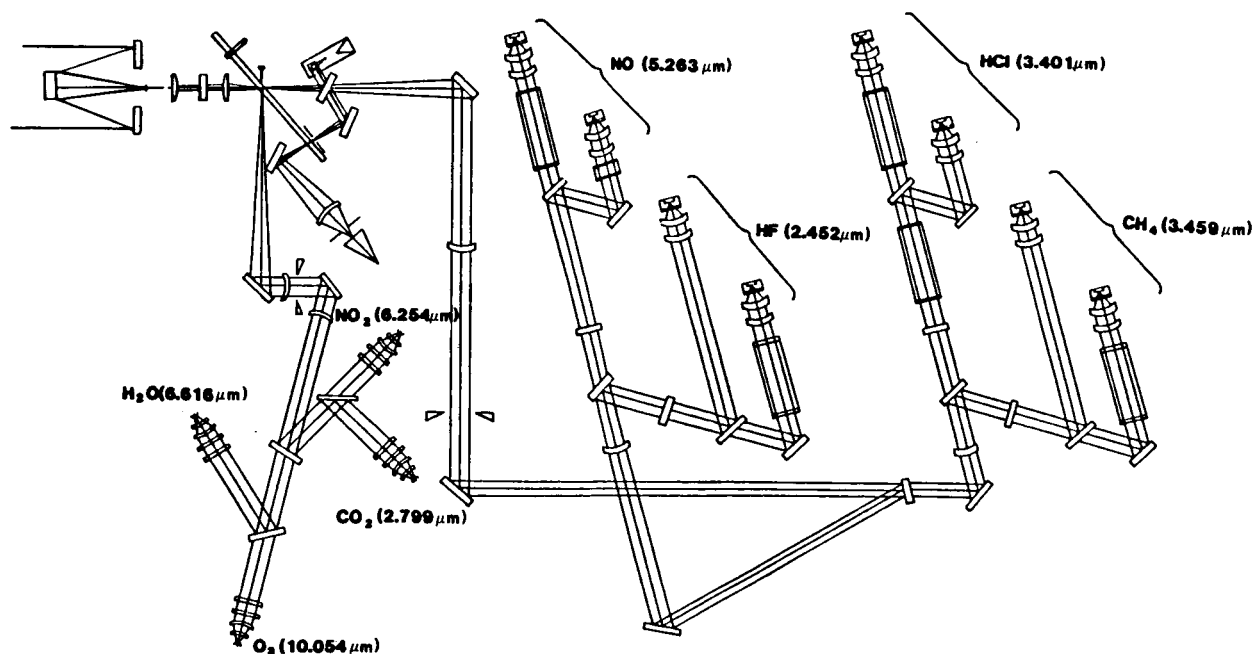


Figure 14.- HALOE schematic.

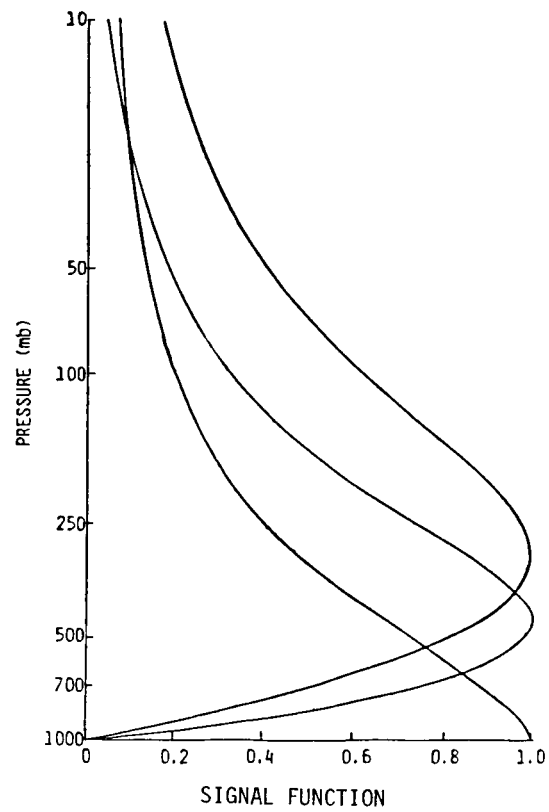


Figure 15.- Signal functions (advanced sounder).

SPECTRAL RADIOMETRY AND TROPOSPHERIC AEROSOLS: REPORT OF PANEL

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INTRODUCTION

The term aerosols, as used here, refers to the haze, smoke, and dust that appear in the troposphere. The term does not refer to the hydrometeors in cumulus and stratus clouds but does include the sulfuric acid-water droplets which are assumed to predominate in the stratospheric aerosol layer. The aerosol properties that have been measured from satellites and those which can be made in the near term (up to 1992) will be reviewed. The capabilities that will exist in the years 1992-2000, with implementation of EOS, will be discussed next. Finally, a few words will be said concerning the potential for aerosol measurements for the decade after 2000.

CURRENT CAPABILITIES

Two satellite methods have been, and are expected to be, available for measuring aerosol properties up to 1992. The first depends on receiving scattered sunlight from a column of the Earth-atmosphere system extending to the ground, and the other method utilizes solar occultation observations of the Earth's limb. The first method consists of measuring the intensity of scattered sunlight in the visible and near-infrared spectrum. The intensity is linearly proportional to aerosol properties such as mass, surface area, and number. Furthermore, those particles that scatter light most strongly are those with diameters approximately equal to the wavelength. Particles that are an order of magnitude smaller, or larger, are ineffective optically--the smaller ones because of their small cross sections and the larger particles because there are not enough of them. A few exceptions can be made, however. If there are enough large particles, such as is the case for Saharan dust, then they are visible at wavelengths much smaller than their particle size. With the exception of dust and, possibly, aerosols derived from the ocean, the available sensors measure properties in the size range between 0.1 and 1 μm .

Several sensors are expected to be available in the near term. These include NOAA's polar orbiters carrying the AVHRR and their geostationary satellites carrying the VISSR. The nadir spatial resolution of these sensors is 1 km. AVHRR scans the entire Earth once each day. VISSR scans the Earth within 60° of the subsatellite point every half hour. The prime purpose of each sensor is to take cloud pictures. As a result, neither sensor is calibrated after launch. Post-launch calibration methods (Griggs, 1984; Fraser and Kaufman, 1986) have been developed, however. Other countries may also continue to deploy geostationary satellites with VISSR-type sensors. Research data will be available from observations made from Landsat and other U.S. and foreign sensors.

Nadir-viewing satellite sensors measure the total vertical burden of the aerosol properties. As a result, validation data are difficult to acquire and do not exist

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generally. The one exception is the aerosol optical thickness. It is linearly proportional to the intensity of the scattered light, at least for the common amounts of aerosol, just as for other aerosol properties previously mentioned. The accuracy of these optical thickness measurements indicates the accuracy of the mass, area, and number measurements also.

The optical thickness measurements are validated by values derived from the solar transmission to the ground. The accuracy of the satellite values is highest when observations are made over a uniform, dark surface such as the ocean. The first extensive measurements were made by Griggs (1984) over many oceanic regions. He compared satellite values with values derived from ground-based measurements of solar transmission (fig. 16). The standard error of estimate is 0.05. Measurements over land are more difficult because of variable surface reflectance, which is about the same or more than aerosol reflectance. Nevertheless, the surface reflectance can be monitored on days with small aerosol thickness. Values of optical thickness measured from a satellite at several locations are compared with ground-based values in figure 17. The good agreement results in the square of the correlation coefficient $r^2 = 0.89$.

Previous values of optical thickness measured from a nadir-viewing satellite were based on measurements in one spectral band. If two or more bands are used, then information about the distribution of physical properties, with respect to size, can be derived. The intensity of light scattered by aerosols relative to the solar spectral irradiance, or aerosol reflectance, depends on the aerosol size distribution. If the particles are much smaller than the wavelength, the aerosol reflectance varies as λ^4 ; if the particles are larger than the wavelength, the reflectance is independent of wavelength. Usually the reflectance varies as λ^{-4} to λ^{-2} . The wavelength dependence of an aerosol size parameter has been derived from satellite measurements, but the results have been validated with only a few independent observations (Griggs, 1984; and Fraser and Kaufman, 1985).

An example of deriving the aerosol albedo of single scattering from Landsat observations of Washington, D.C., during August 1982, is given in figure 18. The reflectances of dark pixels increased from 0.03 to 0.13 with increasing turbidity. The intensity of light from pixels with surface reflectance of 0.18 changed only slightly. If one assumes that the aerosol particles are small and that their absorption cross section is much greater than their scattering cross section, then their albedo of single scattering is 0.9. This value was lower than the satellite value of 0.96 associated with hazes over the eastern U.S. during the summer. In the former case, however, smoke from forest fires over northwestern Canada had drifted over Washington, indicating the presence of soot.

The only remotely sensed clue concerning the chemical composition of the aerosols is the increased size of aerosol particles caused by condensation of water vapor. Then the decrease in optical thickness with respect to wavelength becomes less and, at the same time, the optical thickness increases.

Before global maps of aerosol properties can be derived from nadir-viewing satellite observations, efficient algorithms must be developed. The algorithms have to account for calibration, surface reflectance, and table lookups relating the aerosol properties of models with the intensity of scattered light. The models will depend on the geographical region and season of the year. Extensive tables have to be prepared as a function of solar zenith angle, viewing angles, and wavelength.

Before such nadir-viewing satellite measurements of aerosol properties are accepted, they need to be validated with independent data. Valuable experiments can be made at the surface such as the relation between the mass of sulfate and the scattering coefficient of light (Pierson et al., 1980) or the mass of water condensed on dry particles as a function of humidity. Nevertheless, such experiments are not sufficient since the satellite measures the average properties along a line of sight and the properties vary along the line of sight. Measured vertical profiles are desirable, and they can be obtained by aircraft. Since the aircraft experiments are difficult and expensive, remote measurements from the ground may have to be relied on if several independent measurements yield the same result. For example, measurements of solar transmission and skylight intensity and polarization in many spectral bands may all show the same distribution of aerosol mass with respect to size, giving greater confidence in these satellite data.

The second method of measuring aerosol optical properties utilizes solar occultation of the Earth's limb. This technique has been successfully employed from space with the Stratospheric Aerosol Monitor II (SAM II) flown on Nimbus 7, the Stratospheric Aerosol and Gas Experiment (SAGE) flown on the Application Explorer Mission 2 (AEM-2), and SAGE II flown on the ERBS. In each case, the instrument measures the attenuation of solar radiation through the Earth's limb during spacecraft sunrise and sunset. Such a measurement is essentially self-calibrating, ratioing the attenuated signal through the atmosphere to the unattenuated signal outside the atmosphere to obtain atmospheric transmission profiles.

These data are then inverted to retrieve vertical profiles of aerosol extinction coefficient from cloud tops to approximately 35 km, with a vertical resolution of 1 km and an accuracy of better than 10 percent at the peak of the stratospheric aerosol layer (McCormick et al., 1979; and Mauldin et al., 1985). In cloud-free regions, these profiles can extend down to the Earth surface and do, in fact, reach altitudes of 8 km or lower 50 percent of the time (Woodbury and McCormick, 1983), allowing studies of the upper troposphere as well as the stratosphere. Since these instruments measure the average extinction through the Earth's limb along the spacecraft to Sun view angle, the horizontal resolution of these profiles is $1 \text{ km} \times 200 \text{ km}$. Successive vertical profiles are separated by approximately 24° longitude as the observations sweep through the latitude extremes in 3 to 4 weeks, depending on orbit inclination.

SAM II, launched in 1978 and still operating today, employs a single spectral channel for measurement of aerosol extinction at one wavelength. The Nimbus 7 Sun-synchronous orbit provides geographic coverage from 64° to 80° latitude in both the Northern and Southern Hemispheres. Thus, SAM II is accumulating a relatively large data base on polar aerosols (McCormick, 1981; and McCormick, 1983). SAGE I, launched in 1979, employed four spectral channels for measurement of ozone and nitrogen dioxide in addition to aerosol extinction at two wavelengths. The AEM-2 56° inclination orbit provided geographic coverage from 79° south to 79° north latitude. Thirty-three months of SAGE data were collected before the AEM spacecraft power subsystem developed problems, preventing further instrument operation. However, the almost 3 years of data have been used with the SAM II data set to develop a global upper tropospheric and stratospheric aerosol climatology (McCormick, 1983; McCormick, 1985; Kent and McCormick, 1984).

SAGE II, launched in 1984 and still operating today, employs seven spectral channels for measurement of ozone, nitrogen dioxide, and water vapor, in addition to aerosol extinction at four wavelengths. These data are currently being validated through comparisons with correlative observations such as ground-based and airborne

aerosol lidar backscatter data and balloon-borne in situ particle counters. An example of the preliminary SAGE II aerosol extinction data shown in figure 19 will then be used to study aerosol size and distribution and other aerosol optical properties (Yue et al., 1985).

The ERBS/SAGE II 57° inclination orbit provides geographic coverage from 80° south to 80° north latitude, repeating global coverage every 3 to 4 weeks. Since there are no expendables to be used up, SAGE II could continue to collect data into the early 1990's, providing an important data base for aerosols from the mid troposphere up to 35 km.

PERIOD: 1992-2000

Since the final stage of some important chemical cycles results in the formation of aerosols, the strength of aerosol sources and sinks and their locations are important for understanding atmospheric chemistry. To compute aerosol transport, convergence, and divergence, the aerosol vertical profile and the wind vector profile are required. Lidar can provide the vertical profile of the scattering coefficient. With more than one spectral band, the lidar also gives information about the size dependence of aerosol properties. The lidar data will be relatively infrequent, but the other EOS sensors will provide data on the total vertical burden of aerosol properties on a very small scale. Wind information will be available from standard meteorological data.

Abundant new data rich in spectral information will be available from EOS sensors. A challenge exists to develop algorithms for extracting the information. It is very important to include plans for validating the derived products. Another improvement that EOS will provide is good calibration. Current satellite sensors that measure the intensity of scattered sunlight are either not calibrated or poorly calibrated after launch. The measured reflectance should be accurate within 0.5 percent to measure aerosol properties with accuracy of 10 percent.

The chemical composition of aerosols is exceedingly difficult to measure from satellites. Remote measurements of composition of aerosols in the Earth's atmosphere from the surface of the Earth do not seem to have been made. Laboratory experiments have commenced, however, to identify aerosol species by means of laser backscatter (Goetz et al., 1985). Aerosols in the lower troposphere are screened from satellite observations in much of the spectrum by trace gas absorption. Electronic transitions at energies greater than 3 eV are unobservable because of ozone absorption. Aerosol absorption in the visible and near infrared is dominated by grey absorption. Aerosol vibration bands occur in the 1 to 3 μm window (Bohren and Huffman, 1983) and also in the 8 to 13 μm window (Goetz et al., 1985; C. Prabhakara, NASA Goddard, private communication, 1985). Aerosol spectra will be difficult to measure because of similar spectra from the Earth's surface, numerous absorption lines of water vapor and other trace gases, the small optical thickness of aerosols, and the optical constants are not known for aerosols with mixed composition.

C. Prabhakara (NASA Goddard, private communication, 1985) attempted to extract evidence for the strong silicate band at 1100 cm^{-1} in the Saharan dust as it swept out over the Atlantic Ocean. He used the measurements from the infrared interferometer spectrometer (IRIS) with a spectral resolution of 2.8 cm^{-1} and flown on Nimbus 4. He did not observe the strong Si-O emission near 1100 cm^{-1} , but rather grey emission. Bohren and Huffman (1983) show that such grey emission is expected for dispersed sizes and nonspherical shapes, as are likely for the aeolian dust (Martin and Barber,

1985). This is the only attempt that I know about to identify the chemical composition of aerosols from satellite observations.

To show the difficulty in deriving aerosol spectral information from thermal emission, consider the important aerosol ammonium sulfate. Take as an example measurement of the difference in energy at a satellite from two regions where the difference in ammonium sulfate concentration is $10 \mu\text{g}/\text{m}^3$, as might occur between rural and urban areas (Remsberg, 1978). In order to estimate a relatively strong effect, assume that the aerosol is uniformly mixed in a layer 1500 m deep. Then the total mass of aerosol is $0.015 \text{ g}/\text{m}^2$. The absorption and scattering optical thicknesses for a typical accumulation mode, where sulfates are concentrated, are given in the table below for the minimum, intermediate, and maximum values of extinction in a strong band in an atmospheric window.

A linearized expression for the apparent temperature difference (ΔT) between the rural and urban areas caused by only the ammonium sulfate is

$$\Delta T = \tau_a [T_A - \epsilon_s T_s]$$

where τ_a is the vertical aerosol absorption optical thickness, T_A is the temperature of the aerosol, and ϵ_s and T_s are the emissivity and temperature of the surface, respectively. The maximum $\tau_a = 0.0068$ in the following table, and if the temperature difference between the aerosol and surface emitting as a blackbody is 10K, then $\Delta T = -0.07\text{K}$. Such a small value would seem to be undetectable because of temperature fluctuations caused by the atmosphere and surface, plus radiometer noise.

OPTICAL PARAMETERS OF AMMONIUM SULFATE FOR A STRONG ABSORPTION BAND IN AN ATMOSPHERIC WINDOW*

Wavelength (μm)	Index of Refraction		$C_{\text{ext}} \div \text{mass}$ (m^2g^{-1})	Albedo of Single Scattering	Optical Absorption	Thickness Scattering
	Real	Imaginary				
8.31	1.2183	0.0514	0.042	0.0056	0.0006	0.4×10^{-5}
8.71	1.0227	0.2759	0.234	0.0013	0.0035	0.5×10^{-5}
9.07	1.3909	0.7055	0.458	0.0045	0.0068	3.1×10^{-5}

*The index of refraction is given by Remsberg (1978) for a relative humidity of 41 percent. The logarithm of the sizes is distributed normally with the geometric mean radius = $0.027 \mu\text{m}$ and the geometric standard deviation = 2.16. The mass density is $10 \mu\text{g}/\text{m}^3$, uniformly mixed in layer 1500 m deep.

Lidar detection of the ammonium sulfate aerosols in the $10 \mu\text{m}$ window will be very difficult because they scatter radiant energy very weakly. The aerosol layer for the example given here would scatter at most 3×10^{-5} of the energy from a lidar beam of infrared energy incident on the layer. Since the particles are about 2 orders of magnitude smaller than the wavelength of the $10 \mu\text{m}$ energy, they scatter light according to the Rayleigh phase function. Hence the sulfate layer would scatter backwards per unit solid angle about 5×10^{-6} of the energy incident on it

(neglecting other interactions). An order of magnitude less energy is scattered in the most transparent band near to the strongly absorbing band. To take advantage of such contrast by a differential absorption technique, the numerous absorption lines of water vapor and trace gases have to be avoided.

Satellite mission requirements to measure only aerosol properties are not restrictive. Aerosols are distributed globally with lifetimes of about a week. Strong influxes of aerosols from fires or dust storms can be seen in satellite imagery 10,000 km from their sources. Satellite observations of the entire globe daily would seem to be sufficient.

The local time of the observations requires further study because of clouds. Cumulus clouds are less frequent in the morning over continents. In the absence of clouds, observations with a solar zenith angle of about 60° are favorable, either in the morning or afternoon. For the morning observations, the early morning fog has had time to dissipate. Also, the surface reflection of light from land is less at large solar zenith angle since the intensity of light reflected by the ground varies approximately as the cosine of the solar zenith angle, whereas the intensity of light scattered by the aerosols is not strongly dependent on the solar zenith angle. The major consideration, however, is the limitation caused by clouds, which is more pervasive than just the cumulus problem.

Satellite sensors for EOS will be able to measure the tropospheric aerosol number, surface area, and mass within the size range of 0.1 to 5 μm . Clouds will limit the number of observations that can be made globally each day. Lidar can provide vertical profiles of aerosol physical parameters, but with low coverage. With the use of standard meteorological wind data, horizontal transport, convergence, and divergence can be calculated. The satellite data will be a poor source of information about aerosol chemical composition. The mass fraction of aerosols that absorb radiant energy and also the fraction that is condensed water can be estimated, however.

EOS will provide the basic instruments needed for tropospheric aerosol measurements. A Moderate Resolution Imaging Spectrometer (MODIS) is conceived to measure properties of the land, ocean, and atmosphere in 64 spectral bands, spanning the range between 0.4 and 1.0 μm , plus bands at longer wavelengths. Except for choosing a band at a shorter wavelength of 0.35 μm , measurements from more bands than are needed will be available for deriving aerosol optical properties since they change rather slowly with respect to wavelength and are well correlated. The spatial resolution of MODIS is 1 km at nadir, and it scans the entire Earth every 2 days. Such coverage would be augmented by measurements from other experiments. The aerosol properties can be estimated for the size range 0.1 to a few micrometers. New studies on deriving the aerosol properties from measurements in many bands and the accompanying algorithms are required.

There are currently no plans to continue SAGE II operations or to fly another occultation experiment in this time period. However, because of its relatively low cost, reliability, and capability to measure tropospheric aerosols in addition to stratospheric aerosols, consideration should be given to flying a modified version of SAGE to obtain multiwavelength extinction data for inferring aerosol optical properties and studying the interchange of constituents between the troposphere and stratosphere. Additional spectral channels could be added to obtain further size resolution of aerosol properties, as well as other gas species such as methane. Other improvements could include flying similar instruments on several platforms or tailoring the platform orbit to improve geographic coverage. Stellar and lunar

occultation measurements should also be considered, especially for EOS polar orbits where global coverage could be obtained almost daily by using several stars. The use of a SAGE device would also be important to correcting various passive remote sensors for aerosol effects.

Lidar observations of aerosols from space platforms such as EOS should provide the best vertical resolution data available, with good horizontal resolution (≈ 100 km) along the satellite nadir-viewing track (subsattellite tracks will, however, be 24° longitude apart) and global coverage on a daily basis. Reasonably accurate (20 percent) aerosol backscatter coefficient profiles will be obtainable with 1 to 2 km vertical resolution, whereas the vertical resolution in mapping tropospheric layers such as Saharan dust or other continental plumes should be better than 100 m (Browell et al., 1985). Measurements of total columnar values (optical depth) by other sensors will complement the lidar data set by filling in gaps between subsattellite tracks.

Measurements of the chemical composition from satellites will be minimal or nonexistent. Composition may be inferred from the location of the aerosol source and properties that can be measured--such as absorption and size distribution. In addition, supplementary data from surface and aircraft observations will help to analyze the satellite observations for composition. The difficulties in measuring aerosol spectra that can identify the aerosol are discussed in this report.

One of the critical difficulties is lack of knowledge concerning the optical constants of aerosols, that is, their real and imaginary indices of refraction. Many aerosols consist of a mixture of substances. Their optical constants are not known and are difficult to measure. For continued progress in remote sensing of aerosol properties, their optical constants have to be measured.

PERIOD: 2000-2010

Two significant improvements in aerosol measurements are expected for this period. The first is higher lidar coverage spatially. Additional spectral bands will be available also. The second advance will be the measurement of winds from Doppler shifts in the light scattered from laser beams and also Doppler shifts in emission spectra from the limb of the atmosphere. In addition, studies are in progress to upgrade the geostationary satellites for this period by increasing the number of spectral bands and reducing the instantaneous field of view, thus reducing the cloud contamination problem. If the analyses of previous satellite data continue before this period, then scientists will be prepared to accept the challenges presented by new remote sensing observations.

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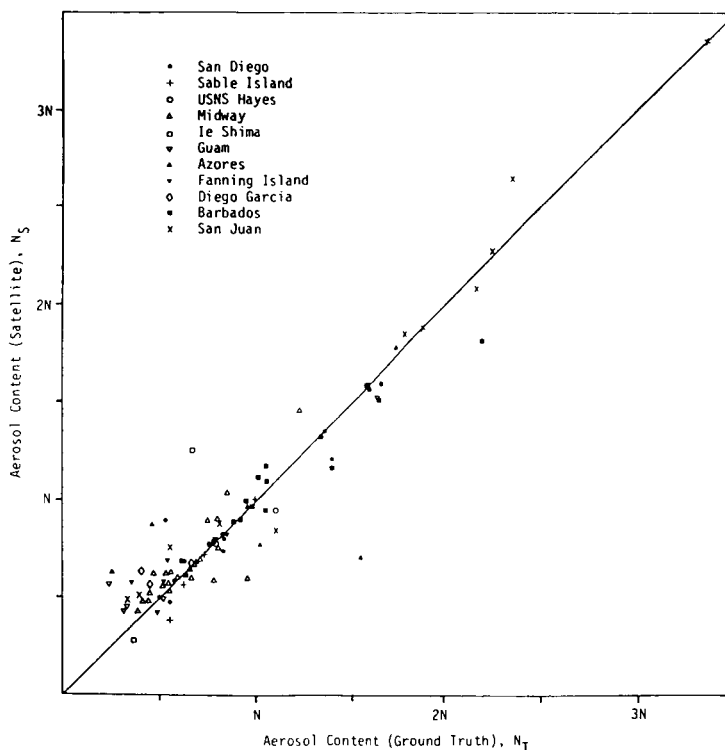


Figure 16.- Satellite measurements of aerosol optical thickness at λ 500 nm by Griggs are compared with ground-based values. One N-unit is equivalent to 0.21 (Toon and Pollack, 1976).

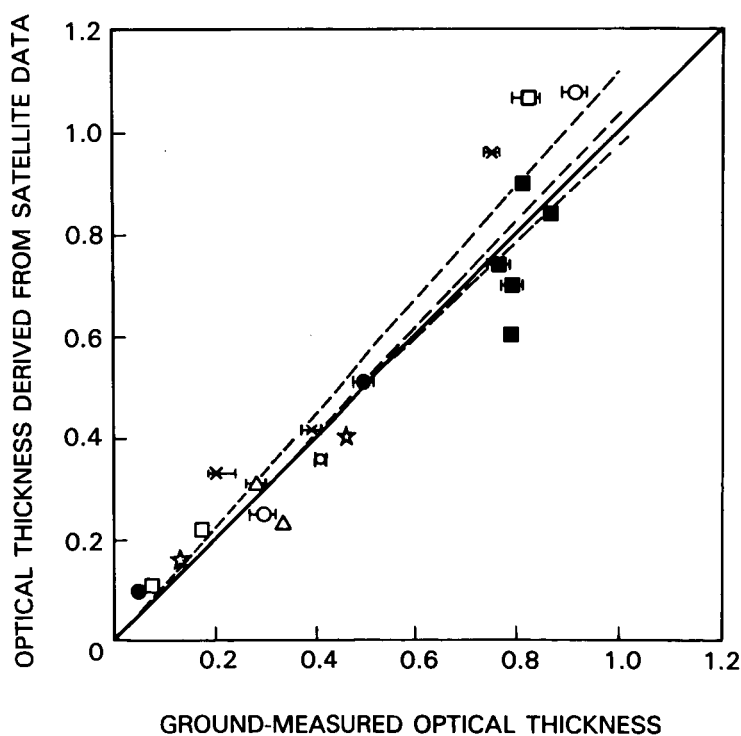


Figure 17.- Comparison of aerosol optical thicknesses measured from a geostationary satellite and the ground for λ 610 nm. The three regression lines were calculated for 1000 sets of data by the bootstrap statistical method and include 34, 50, and 68 percent of those lines (Fraser and Kaufman, 1985a). Some of the ground-based measurements were made by Prospero (J. M. Prospero, private communication, 1982).

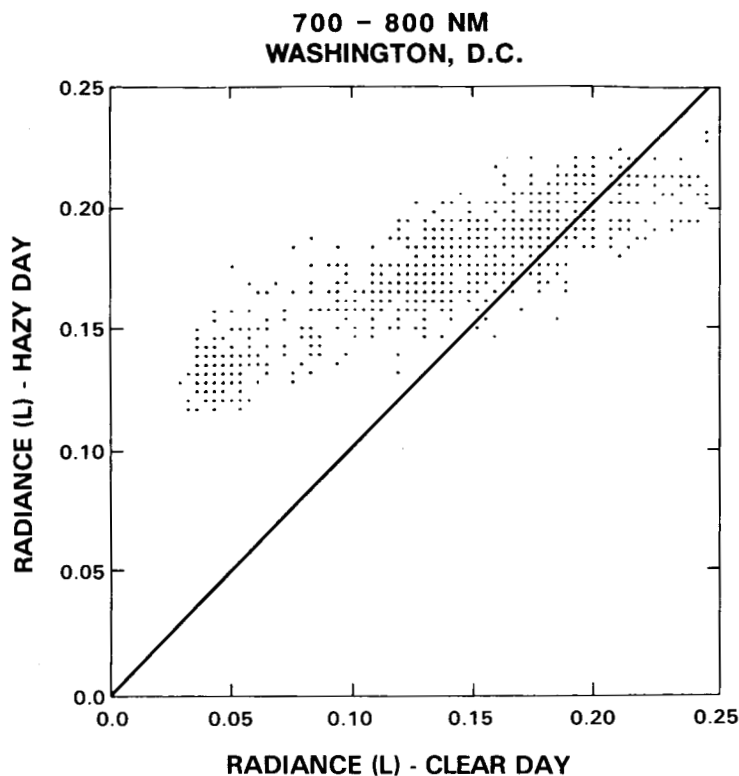


Figure 18.- The change in normalized radiance from a day of low aerosol content to one of high aerosol content in August 1982. The term L is the radiance in the Landsat 700-800 nm band; F_0 is the extraterrestrial spectrum irradiance; and μ_0 is the cosine of the solar zenith angle (Kaufman and Fraser, 1984).

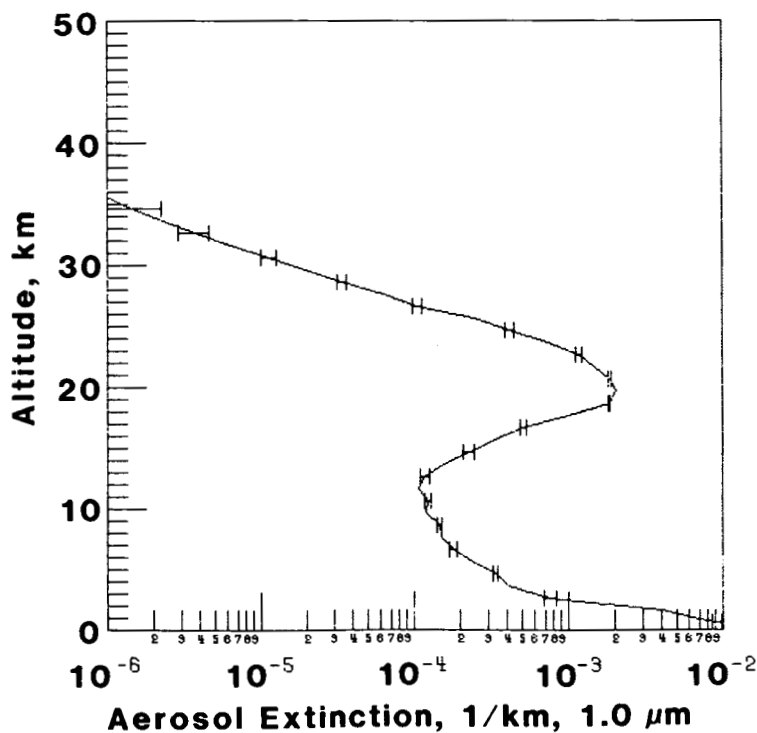


Figure 19.- Aerosol extinction (km^{-1}) profile derived from SAGE II solar occultation measurements at λ 1.02 μm . One sigma error is shown as horizontal bars on the profile.

LIDAR TECHNOLOGY MEASUREMENTS AND TECHNOLOGY: REPORT OF PANEL

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Lidar is ready to make an important contribution to tropospheric chemistry research with a variety of spaceborne measurements that complement the measurements from passive instruments. Over the last two decades, the development of lidar techniques and systems has evolved to the point where it is being used in many different atmospheric investigations from ground, airborne, and balloon platforms. Lidar can now be considered for near-term and far-term space missions dealing with a number of scientifically important issues in tropospheric chemistry. Measurements that can be made by a spaceborne lidar system include distributions of aerosols and gases using the active remote sensing techniques previously discussed. Since there is a scarcity of data on the global distribution of tropospheric species such as O_3 , H_2O , and aerosols, a lidar system in space could make a valuable contribution in conducting tropospheric investigations in conjunction with passive remote measurements of the same or complementary species. In this section, we will address the evolution in the lidar missions from space. Details of these missions are given in table VIII, which is based on the information generated by the Lidar Measurement and Technology Working Group report and on the report by E. V. Browell (both of which are presented in this document).

Three time periods were considered in the evolutionary development. In the near-term period of Space Shuttle flights in 1990 and EOS missions after 1993, lidar measurements of aerosols, H_2O , and O_3 distributions can be made in the troposphere. A single wavelength lidar system operating from 0.5-1.0 μm can provide measurements of aerosol structure through the troposphere with a vertical resolution of ≤ 0.5 km. These data contain information on planetary boundary-layer depth, stable layers in the free troposphere, cloud heights, and other inferred meteorological parameters. A multiple wavelength lidar measurement of aerosols can also provide some information on aerosol size distributions which may be used to identify the type of aerosols being detected. In this time period, a DIAL system may be flown to investigate mid-to-lower tropospheric H_2O globally. The H_2O DIAL measurements will include H_2O column content and vertical profiles with 1 to 2 km resolution. Measurements of O_3 column content in the troposphere and O_3 profiles with 2 km vertical resolution can also be made. DIAL measurements of H_2O and O_3 are made simultaneously with aerosol distribution measurements at the off-line wavelength.

The second period in the evolution of laser remote sensing from space would include possible Space Shuttle flights in the mid 1990's, with an EOS mission by 1997. This requires that the lidar/DIAL technique and technology be ready by about 1992. For this period, we expect to make spaceborne H_2O DIAL measurements with 1 km vertical resolution throughout the troposphere during the day and night. The column

*Prepared panel report.

content of NH_3 can be obtained using an IR DIAL measurement, and temperature can be measured with an O_2 DIAL technique with about 1 km vertical resolution. It is noted that all of the lidar measurement capability noted for the initial period is also available for this period.

The third period, from 2005-2010, includes future DIAL measurements from space of trace species that currently have only been done at elevated levels in the troposphere with low-power DIAL systems. With expected developments in laser systems, it is possible to envision spaceborne DIAL column content measurements of NO_2 , SO_2 , NO , CH_4 , CO , and, possibly, H_2O_2 .

INTRODUCTION: REQUIRED LIDAR DEVELOPMENT AND MEASUREMENTS

The use of a lidar system for the remote sensing of a tropospheric species from space requires advances in laser and detector technology and in supporting spectroscopic and lidar atmospheric measurements. These advances are dependent upon the type of lidar system that is being considered and the time frame for the expected implementation of the lidar or DIAL system for both Space Shuttle and EOS applications. This time frame has been broken into three categories--designated by I, II, and III--which represent, respectively, near term (1986-1992), long term (1990-2000), and those techniques in the far future which will require a significant technology breakthrough to be realized.

Using these criteria, the lidar panel produced two compendiums of their assessment of laser and lidar technology and an approximate listing of the expected time that such lasers would be available for space missions. The first compendium consisted of worksheets that describe the technical details for the laser remote sensing of each atmospheric constituent. The technical issues addressed include laser availability, optical detection sensitivity, and a listing of the most serious technical issues. These worksheets were used to compile a second compendium on the lidar technology assessment, which is a summary of the results from the worksheets. This "summary" assessment is given in the following section.

SUMMARY OF LIDAR TECHNOLOGY ASSESSMENT

Table IX is an assessment of laser availability for space missions based upon the technical data to be given in the worksheets presented.

The time frame for using each type of laser is given for missions on the Space Shuttle and on EOS with low and high duty cycles. For example, water vapor measurements could be conducted using an Alexandrite laser on the Space Shuttle by 1990, EOS (with 10-percent duty cycle) by 1993, and on EOS with long life by 1997. A continuous-wave (CW) CO_2 laser is felt to be able to measure the column content of water vapor from the Space Shuttle and EOS in the same time frames. Aerosols and clouds and Excimer-based O_3 measurements are the only other lidar applications that may be available by 1993 for EOS. By 1997, the measurement of NH_3 may be possible from EOS, using a CO_2 laser system. Long-life DIAL investigations of H_2O , O_3 , and NH_3 may be conducted from EOS in the late 1990's. Measurements of CO , CH_4 , NO , NO_2 , SO_2 , and H_2O_2 from space will be paced by the development of suitable lasers. It is expected that DIAL measurements of these species from EOS will be ready in the 2010 time frame.

It is informative to group these missions by the indicated time frames, and then list the most stressing technical issues for the development of spaceborne lidar systems. These may be expressed as follows.

Phase I: Near-Term Technology Development and Measurements

A. Aerosol Lidar

1. Need high-altitude feasibility demonstration of lidar detection of tropospheric aerosols using Nd:YAG, Excimer, or Alexandrite lasers.
2. Increase efficiency and lifetime of flashlamp pumped Nd:YAG and Alexandrite lasers and discharge Excimer laser.
3. Develop high-efficiency diode pumped solid-state lasers.

B. H₂O DIAL Detection (Column Content)

1. Develop high-resolution Alexandrite laser for DIAL use.
2. Demonstrate DIAL capability at high altitude.
3. Study and develop use of CW CO₂ homodyne (heterodyne) lidar for water vapor detection.

C. O₃ DIAL Detection (Column Content)

1. Demonstrate use of Excimer laser DIAL system at high altitude (>80 km) for tropospheric O₃ detection.
2. Study use of homodyne (heterodyne) CO₂ DIAL for O₃ column content measurements.

Phase II: Long-Term Technology Development and Measurements

A. H₂O Range-Resolved Measurements

1. Increase power and efficiency of Alexandrite laser for H₂O measurements.
2. Study use of Nd:GLASS and Raman shifted technology for upper tropospheric H₂O profiling.
3. Develop diode-pumped tunable solid-state lasers.

B. NH₃ Measurements

1. Develop CW CO₂ heterodyne DIAL for column content NH₃ measurements.

C. NH₃ Temperature Measurements

1. Develop high-resolution Alexandrite laser with precision wavelength control for temperature measurements.

Phase III: Technology Breakthrough Required

A. CH₄, NO, NO₂, and H₂O₂ Measurements

1. These molecules have absorption spectra in spectral regions where current lasers do not have sufficient power for remote sensing purposes and current attempts to reach such wavelengths at high power have not, as yet, been fruitful. However, some new laboratory frequency shifting techniques do hold promise. These include Raman shifting of an Excimer laser and stimulated electronic Raman shifting in cesium vapor using a frequency doubled Alexandrite laser.

B. SO₂

1. SO₂ can be detected in the atmosphere using a UV laser source, but only at very high concentrations such as that emitted by a power plant plume. The optical depth of SO₂ under normal conditions may be too low to permit detection in the ambient troposphere.

C. CO

1. The detection of CO requires a 4.7 μm laser source. Frequency shifting techniques hold promise in reaching this spectral region, but much future work is required before high laser powers will be realized.

TABLE VIII.- PROPOSED EVOLUTION OF LIDAR SPACE MISSIONS

<u>Period</u>	<u>Technology Ready</u>	<u>Shuttle Flight</u>	<u>EOS Flight*</u>	<u>Measurement</u>	<u>Candidate Lasers</u>
I.	1987 \pm 1	1990 \pm 1	1993 \pm 1	Aerosol/cloud distribution and planetary boundary layer (1λ)	Many choices (0.5-1.0 μm)
				Coarse aerosol distribution and tropopause height (3λ)	Nd:YAG (0.35, 0.53, and 1.06 μm)
				H ₂ O - CC and RR (Δz = 2 km, mid-to-low troposphere)	Alexandrite
				O ₃ - CC and RR (Δz = 2 km)	Excimer (Raman-shifted)
II.	1992	1995	1997	H ₂ O (Δz = 1 km through troposphere)	Nd:GLASS (Raman-shifted)
				NH ₃ - CC	CO ₂
				Temperature (Δz = 1 km)	Alexandrite
III.	2000	2005	2010	NO ₂ - CC	Excimer (Raman-shifted)
				SO ₂ - CC	Excimer (Raman-shifted)
				NO - CC	Not identified
				CH ₄ - CC and RR	Not identified
				CO - CC and RR	Not identified
				H ₂ O ₂ - CC	Not identified

* Lidar operated at 10 percent duty cycle, Period I.

CC \equiv Column content

RR \equiv Range resolved (profiles)

$n\lambda$ \equiv n number of lidar wavelengths

Δz \equiv vertical resolution of lidar measurement

TABLE IX.- ASSESSMENT OF LASER AVAILABILITY FOR SPACE MISSIONS

Species	Laser	Time Frame		
		Shuttle*	EOS (10% Duty)	Long Life EOS
H ₂ O	Alexandrite	I	I**	II
	CO ₂ (CW)	I+	I+	II-III
	Ti:Al ₂ O ₃	II	II	III
	Nd:YAG-Dye	II	II	III
	Nd:GLASS	II	II	III
	OPO	II	II	III
O ₃	Excimer	I	I	II
	CO ₂	II	II	II
CO	2X-CO ₂	III	III	III
CH ₄	?	III	III	III
NH ₃	CO ₂	II	II	II
Aerosols	Nd:YAG (2X; 3X)	I	I	I-II
	Excimer + Raman	I	I	II
	Alexandrite	I	I	II
	Ti:Al ₂ O ₃	II	II-III	II-III
	Diode-pumped YAG	II	II-III	II-III
NO	2X-CO ₂	III	III	III
NO ₂	Excimer + Raman	II	III	III
SO ₂	Excimer + Raman	II	III	III
H ₂ O ₂	?	III	III	III

*Shuttle: I--1990; II--1995; III--2005

EOS: I--1993; II--1997; III--2010

**Category I for H₂O column content and lower trop. H₂O profiles (mostly at night)

Category II for lower and upper trop. H₂O profiles day/night

*Column content only

2X ≡ frequency doubled laser output

3X ≡ frequency tripled laser output

WORKSHEETS ON LIDAR TECHNOLOGY

This section presents the worksheets produced by the Lidar Measurements and Technology Working Group. They essentially list the most pressing technical issues for the laser remote sensing of each identified atmospheric constituent from space.

LASER REMOTE SENSING WORKSHEET

SPECIES: **Aerosols**

1. Wavelength: 1.06 μm
 0.53 μm
 0.355 μm
2. Adequate Optical Depth: N/A
3. Any Unique Spectra
 Requirements: None
4. Appropriate Laser Source: Nd:YAG + 2X, 3X

TECHNICAL ISSUES:

- Well demonstrated
 - Probable lifetime, efficiency?
 - Detect at 1.06 μm --limited availability of PMT
 - Size distribution from 3 λ values
-
5. Estimate of Appropriate • Airborne demonstration completed
 Field Demonstration: • LITE (Shuttle) 1989
 6. Estimate of Measurement • 1 λ \rightarrow 50 percent error
 Resolution (Accuracy): • 3 λ \rightarrow 10 percent error

LASER REMOTE SENSING WORKSHEET

SPECIES: Aerosols

1. Wavelength:
 - 0.3 μm
 - 10.6 μm
2. Adequate Optical Depth:
 - Okay at 0.3 μm
 - Little backscatter at 10.6 μm
3. Any Unique Spectra Requirements: None
4. Appropriate Laser Source:
 - Excimer (0.3 μm)
 - CO₂ (10.6 μm)

TECHNICAL ISSUES:

Excimer

- Raman shift to 0.35, 0.5, and 0.6 μm

CO₂

- Need heterodyne detection
- Outgrowth of Doppler lidar
- More complex than 1 μm
- If air is clean, then no signal

5. Estimate of Appropriate Field Demonstration:
 - Excimer--Need airborne demonstration
 - CO₂--Laboratory demonstration complete
6. Estimate of Measurement Resolution (Accuracy): TBD

LASER REMOTE SENSING WORKSHEET

SPECIES: **Aerosols**

1. Wavelength: 0.7 μm
0.5 μm
2. Adequate Optical Depth: N/A
3. Any Unique Spectra Requirements: None
4. Appropriate Laser Source:
 - Alexandrite (0.7 μm)
 - Ti:Sapphire (0.7 μm)
 - Diode Pumped (0.5 μm)
Nd:YAG (2X, 3X)

TECHNICAL ISSUES:

Alexandrite

- Energy Okay (1/2 joule)
- Lifetime?
- Efficiency?
- Possibility of
Doppler/aerosol
spectral resolution

Ti:Sapphire

- 2-5 years development needed

Diode Pumped

- 2-5 years development required

5. Estimate of Appropriate Field Demonstration: 1988
6. Estimate of Measurement Resolution (Accuracy): TBD

LASER REMOTE SENSING WORKSHEET

SPECIES: H₂O

1. Wavelength: 0.72 μm
2. Adequate Optical Depth: Okay
3. Any Unique Spectra Requirements: $\Delta\lambda = 1 \text{ pm}$ lower troposphere spectral purity >99 percent
4. Appropriate Laser Source:
 - YAG Pumped Dye
 - Alexandrite
 - Ti:Al₂O₃

TECHNICAL ISSUES:

YAG Pumped Dye

- Current status power x 5 too low
- Spectral purity problem
- Large size
- Lifetime: 10⁵-10⁶ shots

Alexandrite

- Power okay for CC
- Power low (X3 for range resolved)
- Low efficiency
- Lifetime?
- P \approx 1/2 joule in 2 years

Ti:Al₂O₃

- Low proven power
- Need demonstration tuning and line-width

Need wavemeter development

5. Estimate of Appropriate Field Demonstration: 1988--Alexandrite DIAL system with wavemeter demonstrated in ER-2
6. Estimate of Measurement Resolution (Accuracy): 5 percent CC
10 percent ΔR

LASER REMOTE SENSING WORKSHEET

SPECIES: H₂O

1. Wavelength: 0.94 μm
2. Adequate Optical Depth: Okay
3. Any Unique Spectra Requirements: 0.5 picometer linewidth for upper troposphere
4. Appropriate Laser Source:
 - Dye
 - Nd:GLASS
 - Ti:Al₂O₃ and OPO

TECHNICAL ISSUES:

Dye

- Low power

Nd:GLASS

- Raman shift
- Emerging technology:
slabs (2-5 years)

Ti:Al₂O₃ and OPO

- Requires development
- Raman shift

Detectors: Avalanche photodiode?

5. Estimate of Appropriate Field Demonstration: ~1988 for Dye and Nd:GLASS DIAL measurements
6. Estimate of Measurement Resolution (Accuracy): Aerosol backscatter same as 0.72 μm H₂O

LASER REMOTE SENSING WORKSHEET

SPECIES: H₂O

1. Wavelength: 10.6 μm
2. Adequate Optical Depth: Optical depth?
3. Any Unique Spectra Requirements: TBD
4. Appropriate Laser Source: CO₂

TECHNICAL ISSUES:

- Heterodyne detector required
- (Okay for CW--Column content)
- Use Homodyne

5. Estimate of Appropriate Field Demonstration: TBD
6. Estimate of Measurement Resolution (Accuracy): TBD

LASER REMOTE SENSING WORKSHEET

SPECIES: O₃

1. Wavelength: 0.308 μm
2. Adequate Optical Depth: Okay
3. Any Unique Spectra Requirements: No frequency control; O₃ band is broad
4. Appropriate Laser Source: Excimer (Need ~4Å filter for detection)

TECHNICAL ISSUES:

- o Raman shift for off-line
- o Current powers okay
- o Lifetime?
- o Penetration through stratosphere?
- o Good atmospheric backscatter at 3080 Å

5. Estimate of Appropriate Field Demonstration: Need balloon tests
6. Estimate of Measurement Resolution (Accuracy): TBD

LASER REMOTE SENSING WORKSHEET

SPECIES: O_3

1. Wavelength: $10\ \mu m$
2. Adequate Optical Depth: Okay
3. Any Unique Spectra Requirements: Single Frequency
4. Appropriate Laser Source: CO_2

TECHNICAL ISSUES:

Pulsed CO_2

- Okay for column content
- Possible lifetime problem
- Differential albedo problem
- Penetration through stratospheric O_3 ? (Tune to side of line.)

CW CO_2

- Column content
- No lifetime problem

5. Estimate of Appropriate Field Demonstration: Need balloon tests (35-40 km)
6. Estimate of Measurement Resolution (Accuracy): 10 percent?

LASER REMOTE SENSING WORKSHEET

SPECIES: NH_3

1. Wavelength: $10\mu\text{m}$
2. Adequate Optical Depth: TBD
3. Any Unique Spectra Requirements: May be problem due to small optical depth and shallow layer
4. Appropriate Laser Source: CO_2

TECHNICAL ISSUES:

- Heterodyne detection
- Same status as CO_2 DIAL for O_3 (except no stratospheric burn through problem as with ozone)

5. Estimate of Appropriate Field Demonstration: TBD
6. Estimate of Measurement Resolution (Accuracy): TBD

LASER REMOTE SENSING WORKSHEET

SPECIES: CH₄

1. Wavelength: 7.7 μm
3.3 μm
2. Adequate Optical Depth: TBD
3. Any Unique Spectra
Requirements: TBD
4. Appropriate Laser Source: No current lasers

TECHNICAL ISSUES:

Future Candidate Lasers:

- a. Raman shift Alexandrite
- b. Solid-state tunable laser

5. Estimate of Appropriate
Field Demonstration: TBD
6. Estimate of Measurement
Resolution (Accuracy): TBD

LASER REMOTE SENSING WORKSHEET

SPECIES: CO

1. Wavelength: 4.6 μm
2.3 μm
2. Adequate Optical Depth: Okay
3. Any Unique Spectra Requirements: TBD
4. Appropriate Laser Source: Doubled CO₂ or CO at 4.6 μm

TECHNICAL ISSUES:

Current Lasers Lack Power at 4.6 μm

- 2X CO₂--Need heterodyne detector (L. O. problem)
- Possible CO laser (optical depth?)

No Current Laser at 2.3 μm

Future: Raman shift
(5 Years?)

5. Estimate of Appropriate Field Demonstration: TBD
6. Estimate of Measurement Resolution (Accuracy): TBD

LIDAR MEASUREMENTS FROM SPACE FOR TROPOSPHERIC CHEMISTRY INVESTIGATIONS:
SUMMARY OF WORKSHOP OVERVIEW PRESENTATION

E. V. Browell

ABSTRACT

Over the past decade, NASA has played a lead role in defining the scientific objectives and technology requirements for spaceborne lidar investigations of the atmosphere. This paper presents an assessment of the potential for conducting lidar measurements from space for investigations that pertain specifically to tropospheric chemistry. In this paper, a description of potential lidar measurement techniques is given, and the scientific requirements for tropospheric chemistry are reviewed. The current status of airborne lidar measurements of aerosols, O_3 , and H_2O is discussed, and a brief description of the evolution of lidar technology to space is given. Also, the measurement of tropospheric gases with a spaceborne lidar system is evaluated for a wide range of gas species. From this general assessment, it appears feasible to measure aerosols, H_2O , O_3 , NH_3 , CO , CH_4 , NO_2 , atmospheric pressure and temperature, and winds with a lidar from space provided that the appropriate laser and receiver technology is available. For the mid-1990's, it is expected that lidar technology will be available for the measurement of aerosols, H_2O , and O_3 from a space platform.

INTRODUCTION

Lidar has been used extensively since the early 1960's for making measurements of various properties of the Earth's atmosphere. These include molecular and aerosol backscattering, gas concentration profiles, wind velocities, and atmospheric waves. The development of lidar in the early years was primarily concerned with laboratory and ground-based systems. Measurements with airborne lidar systems were not made until the early 1970's. By 1975, the lidar technique was well proven and consideration was being given to developing a shuttle-borne lidar system. A lidar system is now undergoing development to demonstrate the basic measurements of aerosol and cloud distributions from space.

There are four primary atmospheric lidar processes or techniques: elastic scattering, Raman scattering, resonance fluorescence, and differential absorption. Lidar measurements utilizing elastic backscattering have focused primarily on investigations of molecular density, aerosol and cloud distributions, and winds (e.g., Fiocco and Grams, 1966; Lane et al., 1971; Bilbro and Vaughan, 1978). Lidar systems detecting Raman-shifted frequencies have generally been limited to measurements of gases having high mixing ratios, such as water vapor, at relatively short ranges of 1 to 4 km at night (Cooney, 1971; Melfi and Whiteman, 1985). Resonance fluorescence has been an important technique used to measure sodium, potassium, and atmospheric waves in the upper atmosphere (e.g., Bowman et al., 1969; Megie et al., 1978; and Chanin and Hauchecorne, 1981). The Differential Absorption Lidar (DIAL) technique focuses on the simultaneous measurement of aerosol and gas concentration profiles, and it is the primary lidar technique for use in tropospheric gas measurements from space. This technique requires the near-simultaneous transmission of two wavelengths, usually by two tunable lasers. One of the lasers is tuned "on" the peak of an absorption line of the species to be measured, and the other is tuned "off" the absorption peak to a nearby wavelength. The two laser beams are backscattered from the

atmosphere to a collocated receiver where the signals are digitized as a function of range. The average gas concentration n between range R_1 and R_2 can be calculated using the expression (Schotland, 1974)

$$n = \frac{1}{2(R_2 - R_1)(\sigma_{\text{on}} - \sigma_{\text{off}})} \ln \frac{P_{\text{off}}(R_2) P_{\text{on}}(R_1)}{P_{\text{off}}(R_1) P_{\text{on}}(R_2)}$$

where $\sigma_{\text{on}} - \sigma_{\text{off}}$ is the absorption cross section difference between the on and off wavelengths, and $P_{\text{off}}(R)$ and $P_{\text{on}}(R)$ are the detected backscattering signals received from range R for the on and off wavelengths, respectively.

As shown in figure 20, the development and application of lidar techniques have seen a steady growth over the last two decades. Early ground-based DIAL systems measured atmospheric H_2O and aerosols (Schotland, 1966; Browell et al., 1979); SO_2 in power plant stack plumes (Hoell et al., 1975; Browell, 1982); and O_3 in the lower atmosphere (Pelon and Megie, 1982). Recently, an advanced airborne DIAL system was developed and has been used to measure O_3 , H_2O , and aerosols in the troposphere (Browell et al., 1983). Studies of spaceborne lidar measurements and systems can be traced back to the early 1970's. The most comprehensive study of the science objectives and measurement capabilities of a spaceborne lidar system was published by NASA in 1979 (NASA, 1979). These activities led to the development of an autonomous DIAL system for the measurement of H_2O and aerosols from a high-altitude ER-2 aircraft as a precursor to a spaceborne DIAL system.

A working group was formed by NASA in 1978 to identify the major measurement requirements in tropospheric chemistry and to develop the scientific rationale and recommended research activities to be conducted by NASA to increase our understanding of the troposphere (NASA, 1981). A summary of the measurement requirements identified by that group is given in table X. A minimum of two gas concentration measurements in the troposphere, one across the boundary layer and one across the free troposphere, was determined to be necessary for most tropospheric chemistry applications. Listed in table X are the minimum detectable gas concentration levels that would be required. The measurement of atmospheric state variables was not identified to be of strong importance in tropospheric chemistry applications, and thus, relatively low accuracies were needed for the measurement of these parameters. Information on aerosols was cited as important for the understanding of heterogeneous chemistry, atmospheric structure, and atmospheric transport.

This paper discusses the status of current airborne lidar measurements of aerosols, O_3 , and H_2O and the planned evolution of lidar technology to space. A general evaluation of gas measurements with lidar from space is presented. Future developments in lidar techniques and measurements that are suitable for space applications are assessed, and technology requirements for future spaceborne lidar systems for tropospheric measurements are presented.

AIRBORNE LIDAR MEASUREMENTS OF AEROSOLS, O_3 AND H_2O

An advanced airborne DIAL system has been developed at the NASA Langley Research Center for the investigation of aerosols, O_3 , and H_2O in the troposphere (Browell et al., 1983). A schematic of the airborne DIAL system mounted in the NASA Wallops

Electra aircraft is shown in figure 21. In this system, two frequency doubled Nd:YAG lasers are used to pump two high conversion efficiency dye lasers. The on and off wavelengths are produced in sequential laser pulses with 100 to 250 μ s separation. Backscattered lidar returns at the two wavelengths are sequentially detected by a photomultiplier tube, digitized, and stored on high-speed magnetic tape. Gas concentrations and aerosol backscattering profiles are calculated for each measurement in real time by a minicomputer. The selection of the lasers was dictated by three basic requirements: tunability, high power, and reliability. The dye laser is easily tuned with different dyes over a wide selection of wavelengths from 400 to 1200 nm, and the output can be frequency doubled into the UV. The performance parameters of the airborne DIAL system operating in the UV and near-IR are listed in table XI.

Examples of airborne DIAL data are shown in figure 22 to illustrate some of the capabilities of the system. Signal returns from the UV (300 nm) and visible (600 nm) channels are displayed at the top of the figure to show the relative contrast resulting from increased aerosol backscattering in the atmospheric boundary layer. The range-corrected visible channel is shown at the lower left, and a gray scale picture made using these data is shown at the lower right. Each vertical line of the gray scale represents a laser shot, and the darker regions represent greater atmospheric backscattering, or aerosol concentration. This gray scale clearly defines the boundary layer height and presence of clouds (dark regions followed by absence of return signal from lower altitudes). This type of aerosol data can provide information on widely varying atmospheric conditions including aerosol layering and transport, clouds, condensation levels, and topographic features. Figure 23 presents an intercomparison of DIAL O_3 measurements with in situ measurements made onboard a small aircraft spiraling in the vicinity of the Electra. The intercomparison shows that: (1) the remote lidar measurements and in situ measurements are in excellent agreement, (2) the DIAL measurements have 10 percent or less measurement uncertainty for the spatial resolution shown, and (3) the DIAL measurements can be made with high vertical (210 m) and horizontal (6 km) resolution.

Airborne DIAL measurements have been used to study the correlation between spatial distributions of O_3 and aerosols in the troposphere and lower stratosphere. Figure 24 shows O_3 measurements by the DIAL system in the zenith-viewing mode compared with in situ observations. Profiles of potential temperature and ozone measured by an ozonesonde are presented in the figure along with DIAL O_3 data. The location of a change in slope of the potential temperature profile (≈ 11 km) defines the height of the tropopause. The O_3 profiles show layering of O_3 in the lower stratospheric regions, and there is good agreement between the DIAL and in situ measurements. Further, other DIAL measurements show that this O_3 layering persisted along the flight path of the aircraft. This indicates the ability of an airborne DIAL system to map O_3 layers in the troposphere and lower stratosphere and to study stratosphere and troposphere exchanges.

When the airborne DIAL system is used to study H_2O profiles, the on-line laser wavelength is tuned to coincide with the peak of a H_2O absorption line in the 720 nm region. An intercomparison between the DIAL H_2O measurements and in situ observations is shown in figure 25. The good comparison between these two observations shows the ability of the DIAL system to make H_2O measurements in the lower troposphere. This capability was used for the first time in the fall of 1982 for the study of marine boundary layer characteristics in the vicinity of the Gulf Stream Wall (Browell et al., 1984).

EVOLUTION OF LIDAR TECHNOLOGY FOR SPACE

There are three lidar development programs within NASA that are directly aimed at providing the technological base for placing a DIAL system on a free-flying space platform by the mid 1990's. The LASE (Lidar Atmospheric Sensing Experiment) and LITE (Lidar In-Space Technology Experiment) projects address important phases of the spaceborne lidar development program. The LASA (Lidar Atmospheric Sounder and Altimeter) system is the near-term goal for a lidar system operating on a polar orbiting platform. This would be part of the NASA EOS Program.

The LASE system is being developed as a high altitude (16-21 km), autonomous DIAL system for operation on a NASA ER-2 (Extended Range U-2) aircraft. It will initially operate in the 727-nm wavelength region to measure H₂O profiles in the lower troposphere, and at a later stage it will be used in the 940-nm wavelength region to measure H₂O profiles in the upper troposphere. In addition, atmospheric pressure and temperature determinations will be made using DIAL O₂ measurements in the 760-nm wavelength region. Figure 26 shows the results of our simulations to determine the random errors associated with H₂O measurements with the LASE system. The random error at a specific altitude depends upon the absorption strength of the H₂O line being used, with the stronger lines being more suitable for higher altitude regions. Clearly the random errors for night operation and for a horizontal resolution of 20 km and a vertical resolution of 200 m would permit a 5- to 10-percent measurement uncertainty in a specific altitude region. Our analysis shows that systematic errors could contribute a significant amount to the uncertainty of these measurements. The systematic errors can be caused by: (1) interaction of the laser linewidth with the altitude dependent H₂O absorption linewidth, (2) Doppler broadening of backscattered light by air molecules, (3) accuracy in the positioning of the laser line with respect to the H₂O line, (4) uncertainty in knowledge of the laser wavelength, and (5) the broadband laser energy component outside the dominant laser on-line emission. Our simulations indicate that with proper controls on the laser output and by folding in model atmospheric temperature and pressure information, the systematic errors could be limited to a few percent (Ismail et al., 1984). The LASE system would be capable of making measurements on long-range flights (>4500 km) during both day and night background conditions, and because of its unique autonomous mode of operation, the LASE system would be a precursor to the development of a spaceborne lidar system. LASE is expected to make its initial flight in late 1988.

The development of spaceborne lidar systems is contingent on the ability of lasers to operate in a space environment. NASA is taking a step in the direction of developing an operational laser capability for space with the LITE project. The objective of LITE is to develop the technology base and measurement techniques necessary to operate a solid-state laser in a spaceborne lidar system. The approach being used is to space-harden existing lidar technology with minimum change. The initial experiment will utilize a frequency doubled and tripled Nd:YAG laser in a lidar system with simultaneous measurements at three wavelengths. The construction will incorporate easily modified modular concepts, and the experiments will focus on aerosol, cloud, and atmospheric density measurements. LITE is scheduled to fly on the Space Shuttle in 1989.

The next program following LITE to incorporate a lidar system in space will most likely be the EOS Program scheduled for the middle 1990's time period. A lidar system for EOS is being proposed, and it is called the LASA instrument. The science objectives for LASA cover many disciplines, including hydrology, altimetry, geodynamics, aerosol, and cloud studies. The list of lidar measurements proposed for LASA is

shown in table XII. The 14 listed items have all been demonstrated in the laboratory, and most have been demonstrated in both ground-based and low-altitude airborne systems. LASE data from the ER-2 aircraft will further demonstrate high-altitude lidar measurement capabilities. Using the technology base developed from the LASE and LITE programs, LASA should be successful in gathering large quantities of data on a global basis to make a significant step forward in the scientific understanding of our atmosphere.

GAS MEASUREMENTS WITH LIDAR FROM SPACE

The accuracy of a gas concentration measurement with the DIAL technique is dependent upon the absorber thickness across the region where the average gas concentration is to be determined. Figure 27 shows the sensitivity in the error in optical depth determination to the absorber optical depth for various transmittance measurement errors. For small absorber optical depths, the error in the determination of the optical depth decreases with increasing absorber optical depth. An optimum absorber optical depth is reached around a value of 1 (note that this analysis is for a single-pass measurement), and at higher optical depths the error begins to increase precipitously due to loss of signal. Thus, the measurement of a gas using the DIAL technique depends upon the expected gas concentration, the available gas absorption cross sections, and the range over which the absorption is to be measured.

Table XIII lists the mean mixing ratios for the tropospheric gases identified in Table X. Also given in table XIII are the column burdens for the same gases over the entire troposphere. To evaluate the ability of a DIAL system to make a measurement of a gas from space, we must next look at the available absorption cross sections for these gases. In table XIV, the gases from table XIII are listed in order of decreasing mixing ratio in the lower troposphere. For each molecule, the absorption wavelength region is given with an estimate of the maximum absorption cross section for that region (weak lines are not usually difficult to find in the same region). Since the DIAL measurement actually involves a double pass across any altitude segment, estimates of the two-way optical are given across a 1-km boundary layer and across the entire troposphere.

From the information provided in table XIV and from the results of other spaceborne lidar experiment simulations not presented here, the following measurements have been identified as potential objectives for a spaceborne DIAL system, providing the laser and receiver technology are available: (1) profile measurements of H_2O , O_3 , CO , CH_4 , and possibly NH_3 ; (2) tropospheric column content measurements of NO_2 and possibly NO and SO_2 with very low-horizontal resolution; (3) atmospheric state parameter measurements to the required accuracy and direct measurements of winds; and (4) aerosol distribution measurements for information on atmospheric structure and meteorological parameters, and multiple-wavelength aerosol measurements for information on aerosol composition and mass loading.

An example is given in figure 28 of the accuracy for obtaining H_2O profile measurements using absorption lines of different strengths in the 727 nm wavelength region. A vertical resolution of 1.0 km and a horizontal resolution of 100 km were used in these calculations with a laser energy of 500 mJ at 10 Hz and a 1.25-m diameter telescope receiver system. It can be seen that the weaker lines optimize at a lower altitude, and that to provide altitude coverage from the surface to the upper troposphere, the DIAL measurement requires the use of at least two absorption lines.

With the resolution shown, the H_2O profiles measurement could have a random error of $<10\%$ below 5 km altitude and $<15\%$ from 3-10 km altitude. These measurement uncertainties can be further reduced by using a larger vertical and/or horizontal resolution.

STATUS OF DIAL MEASUREMENTS SUITABLE FOR SPACE APPLICATIONS

Table XV summarizes the status of DIAL measurements that pertain to eventual remote sensing of tropospheric parameters from space. The gas species or atmospheric parameter is listed along with the nominal wavelength region for the lidar measurement. In all cases, the theoretical feasibility for an atmospheric lidar measurement has been shown. The atmospheric measurements from a laboratory environment have also been demonstrated from most of the parameters. Some of measurements were made in enhanced gas concentrations in plumes, in an urban environment, or over a roadway. The others were made in the ambient rural or remote troposphere. Fewer field measurements have been made with mobile or portable ground-based lidar systems. Most of these systems have been used for pollution studies of SO_2 or in tracking plumes using aerosols as a tracer. Lidar measurements from aircraft are indicated along with the year the first demonstration was conducted. As can be seen from table XV, all of the activity in airborne DIAL measurements has been aimed at O_3 and H_2O over the last 6 years. Table XVI presents an estimate of where airborne DIAL measurements are going in the near future (<2 years) and beyond. Also indicated is the type of laser or wavelength region that is expected to be used in the measurement. In the near future O_3 will be measured with an excimer-based DIAL system as will SO_2 in power plant plumes. Atmospheric H_2O , pressure, and temperature will be measured using Alexandrite lasers in an airborne DIAL system. Beyond the next two years, DIAL measurements of CH_4 , CO , NH_3 , NO_2 , and NO depend upon the development of appropriate lasers and DIAL systems in the wavelength regions shown.

The general characteristics needed for spaceborne lasers and a list of potential tunable lasers for spaceborne lidar applications are given in table XVII. Most spaceborne lidar applications require tunable laser wavelengths with high average power. The characteristics given in table XVII are for a free-flying satellite that may only be visited every 12 to 24 months. The types of tunable lasers given in the table are examples of general laser types that are currently under development and are potential candidates for future spaceborne lidar missions.

SUMMARY

This paper has discussed the evolution of lidar systems for tropospheric observations. Measurements of aerosols, O_3 , and H_2O made with the NASA LaRC airborne DIAL system were presented as examples of the types of data that could be obtained from a spaceborne lidar system. An autonomous DIAL system under development by NASA for the high-altitude ER-2 aircraft was described. Future spaceborne lidar systems, LITE and LASA, were discussed, as well as the requirements for future spaceborne laser systems. It is clear that the successful development of spaceborne lidar systems depends critically on solving the laser technology challenges of improved laser efficiency and long lifetimes.

Spaceborne lidar can potentially contribute to tropospheric chemistry investigations by making profile measurements of H_2O , O_3 , CO , CH_4 , and possibly NH_3 . Also, it may be possible to measure the tropospheric column content of NO_2 and possibly SO_2

with low horizontal resolution. Winds and atmospheric state variables could also be provided by lidar measurements. Aerosol and cloud characterization can be obtained with a multiple-wavelength lidar system. All of these measurements require the appropriate laser and receiver technology for long-term space applications. By the mid-1990's a spaceborne DIAL system could be placed in orbit to investigate tropospheric profiles of aerosols and H_2O , or possibly investigate low vertical resolution (>3 km) measurements of tropospheric O_3 . The information obtained by these first systems would make an important contribution to tropospheric chemistry investigations. By the year 2000, laser and lidar technology will have advanced to the point where the remote measurement of many other species would be possible. Lidar is an important area of development because there are no other remote sensing techniques that can obtain high vertical resolution (<2 km) measurements of atmospheric parameters and gases in the lower atmosphere from space.

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TABLE X.- PRELIMINARY MEASUREMENT REQUIREMENTS IN TROPOSPHERIC CHEMISTRY*

Resolution - Vertical: Boundary Layer & Free Trop. Minimum
Horizontal: ≤ 200 km

Species (Accept. Detect.) - O_3 (10 PPB), CO (50 ppb), CH_4 (1.5 ppm) NMHC (10 ppt),
 H_2CO (0.10 ppb), NO (10 ppt), NO_2 (10 ppt),
 HNO_3 (10 ppt), OH ($5 \times 10^5 \text{ cm}^{-3}$), HO_2 (10^8 cm^{-3}),
 H_2O_2 (1.0 ppb), H_2O (100 ppm), NH_3 (0.10 ppb),
 SO_2 (50 ppt), H_2S (5 ppt), CH_3SCH_3 (5 ppt), CS_2 (5 ppt),
COS (50 ppt)

State Variables - Temp. ($\pm 5K$), Press. ($\pm 10\%$), Cloud Cover and Height, Freq. and
Dist. of Lightning

Aerosol Measurements - Optical Properties Under Flight Track, Chemical Composition,
Size Distribution

*Adapted from NASA, 1981

TABLE XI.- AIRBORNE DIAL SYSTEM CHARACTERISTICS

Transmitter:

Two Pump Lasers -- Quantel Model 482

Pulse Separation -- 100 μs

Pulse Energy -- 350 mJ at 532 nm

Repetition Rate -- 10 Hz

Pulse Length -- 15 ns

Two Dye Lasers -- Jobin Yvon Model HP-HR

	UV (near 300 nm)	Near-IR (near 720 nm)
Fundamental Dye Output Energy	157 mJ pulse near 600 nm	63 mJ pulse
Doubled Dye Output Energy	47 mJ/pulse near 300 nm	--
Transmitted Laser Energy	40 mJ/pulse near 300 nm and 80 mJ/pulse near 600 nm	50 mJ/pulse
Laser Linewidth	<4 pm	<2 pm

Receiver:

Area of Receiver	0.086 m ²	0.086 m ²
Receiver Efficiency to PMT	28%	29%
PMT Quantum Efficiency	29%	4.8%
Total Receiver Efficiency	8.1%	1.4%
Receiver Field of View	2 mrad	2 mrad

TABLE XII.- DEMONSTRATED LIDAR MEASUREMENTS FOR LASA

	Lab	Ground-Based	Low Alt. Airborne	High Alt. Airborne	LASA Sci. Objectives
1. Altimetry	X	X	X	-	2
2. Retro-Ranging	X	X	-	-	3
3. Cloud Top Height	X	X	X	X	1
4. PBL* Height	X	X	X	X	1
5. Strat. Aero.	X	X	X	-	4
6. Cloud Prop.	X	X	X	X	4
7. Trop. Aero.	X	X	X	X	4
8. H ₂ O Column	X	X	X	A	1
9. Surface Pres.	X	-	-	-	5
10. O ₃ Column	X	X	X	-	5
11. H ₂ O Profile	X	X	X	A	1
12. Pres. Profile	X	X	-	-	5
13. Temp. Profile	X	-	-	-	5
14. O ₃ Profile	X	X	X	-	5

LASA Science Objectives: 1 Hydrology; 2 Altimetry; 3 Geodynamics;
4 Aerosols and Clouds; 5 Other

A: LASE Measurements (1988)

*Planetary Boundary Layer

TABLE XIII.- TROPOSPHERIC GAS ABUNDANCES

Species	Mean Mixing Ratio in Lower Trop.*	Nominal Tropospheric Column Burden** (ppm-m)
O ₃	50 ppb	230
CO	90 ppb	800
CH ₄	1.65 ppm	1.1 x 10 ⁴
NMHC* (C ₂ to C ₁₂)	> 1 ppb	-
H ₂ CO	-	0.14
NO	5 ppt	2.0
NO ₂	10 ppt	1.7
HNO ₃	50 ppt	23
OH	0.05 ppt	0.01
HO ₂	1 ppt	0.12
H ₂ O ₂	500 ppt	-
H ₂ O	1300 ppm	4.1 x 10 ⁷
NH ₃	10 ppb	20
SO ₂	100 ppt	0.81

*NASA, 1981

**Alvarado et al., 1982

TABLE XIV.- TROPOSPHERIC GAS ABSORPTION PROPERTIES

Species	Wavelength Region (μm)	Absorption Cross Section ($\text{cm}^2/\text{Molecule}$)	Est. of Maximum Two-Way Optical Depth	
			Across 1 km B.L.	Across Troposphere
H_2O	0.72	6×10^{-23}	0.42	13
	0.94	2×10^{-21}	14	---
	1.23	4×10^{-20}	280	---
	6.3, 2.7	1×10^{-18}	7000	---
CH_4	7.7, 3.33, 3.25	1×10^{-18}	8.9	---
CO	4.6	2×10^{-18}	0.97	8.6
	2.33	2×10^{-20}	0.010	0.086
O_3	10	1×10^{-19}	0.027	0.12
	0.29	9×10^{-19}	0.24	1.07
NH_3	10	1×10^{-18}	0.05	0.1
SO_2	4.0	3×10^{-20}	1.6×10^{-5}	1.3×10^{-4}
	0.3	1×10^{-18}	5.4×10^{-4}	.0044
NO_2	0.44	3×10^{-19} ($\Delta\sigma$)	1.6×10^{-5}	0.0028
NO	5.4, 5.2	1×10^{-18}	2.7×10^{-5}	0.011

TABLE XV.- STATUS OF DIAL MEASUREMENTS SUITABLE FOR SPACE APPLICATIONS

Gas Species/ Atm. Param.	Nominal Wavelength Range (μm)	Theoretical Feasibility	Atm. Mea. from Lab	Field Mea.	A/C Mea.
O_3	0.3	X (RR)	X (RR)	X (RR)	1980 (RR)
	9.5	X (CC & RR)	X (CC)	X (CC)	1979 (CC)
H_2O	0.72	X (RR)	X (RR)	X (RR)	1982 (RR)
	0.94/1.23	X (RR)	--	--	
	1.8	X (CC)	X (CC)	--	
	10.3	X (CC & RR)	X (CC & RR)	X (CC)	1982 (CC)
CH_4	3.5	X (CC)	X (CC)	--	
CO	5.0	X (CC)	X (CC-Roadway)	--	
NH_3	10.3	X (CC)	X (CC)	--	
SO_2	0.3	X (RR)	X (RR-Plume)	X (RR-Plume)	
NO_2	0.44	X (RR)	X (RR-Plume, CC-Urban)	X	
	--	--		--	
NO	5.3	X (CC)	X (CC-Roadway)	--	
Press. (O_2)	0.76	X (RR)	X (RR)	X	
Temp. (O_2)	0.78	X (RR)	X (CC)	X	
Aerosol Dist.	0.3/0.6/1.0	X	X	X	1968
Aerosol Char.	0.3/0.6/1.0	X	X	X	1984
Winds (Doppler)	10.6	X (Profile)	X	X	1981

TABLE XVI.- AIRBORNE LIDAR TROPOSPHERIC MEASUREMENTS

<u>Demonstrated</u>	<u>Near Future (\leq 2 Years)</u>	<u>Future (\geq 2 Years)</u>
O ₃ (Nd:YAG-DYE; CW-CO ₂)	O ₃ (Excimer)	O ₃ (Pulsed CO ₂)
H ₂ O (Nd:YAG-DYE; CW & Pulsed CO ₂)	H ₂ O (Alex.)	H ₂ O (0.94/1.23 μ m)
SF ₆ Tracer (CW-CO ₂)	SO ₂ (Excimer)	CH ₄ (3.3 μ m)
Aerosol & Cloud Dist. (Nd:YAG)	P (Alex.)	CO (4.6 μ m)
Aerosol Discrim. (N λ Nd:YAG-DYE)	T (Alex.)	NH ₃ (10.3 μ m)
P (2 λ Nd:YAG)	--	NO ₂ (0.44 μ m)
Winds (CW & Pulsed-CO ₂ Doppler)	--	NO (5.4 μ m)

TABLE XVII.- SPACEBORNE LASERS

<u>General Characteristics</u>	<u>Tunable Laser Types</u>
High Power (1 - 20 W)	Alexandrite (720 - 770 nm)
Wavelength Tunable (10 - 40 cm ⁻¹)	Co:MgF ₂ * 1.6 - 2.3 μ m
Narrow Linewidth (<0.02 cm ⁻¹)	Ti:Sapphire (700 - 850 nm)
Long Lifetime (12 - 24 mo)	Emerald (751 - 759 nm)
Ruggedized (Shuttle Launch; Space Environ.)	Nd:Glass*
Reduced Complexity and Modular	Excimer*
Capable of Meeting Eye Safety Criteria	CO ₂ (Line Tunable and Isotope Broadened)

*Raman Shifted

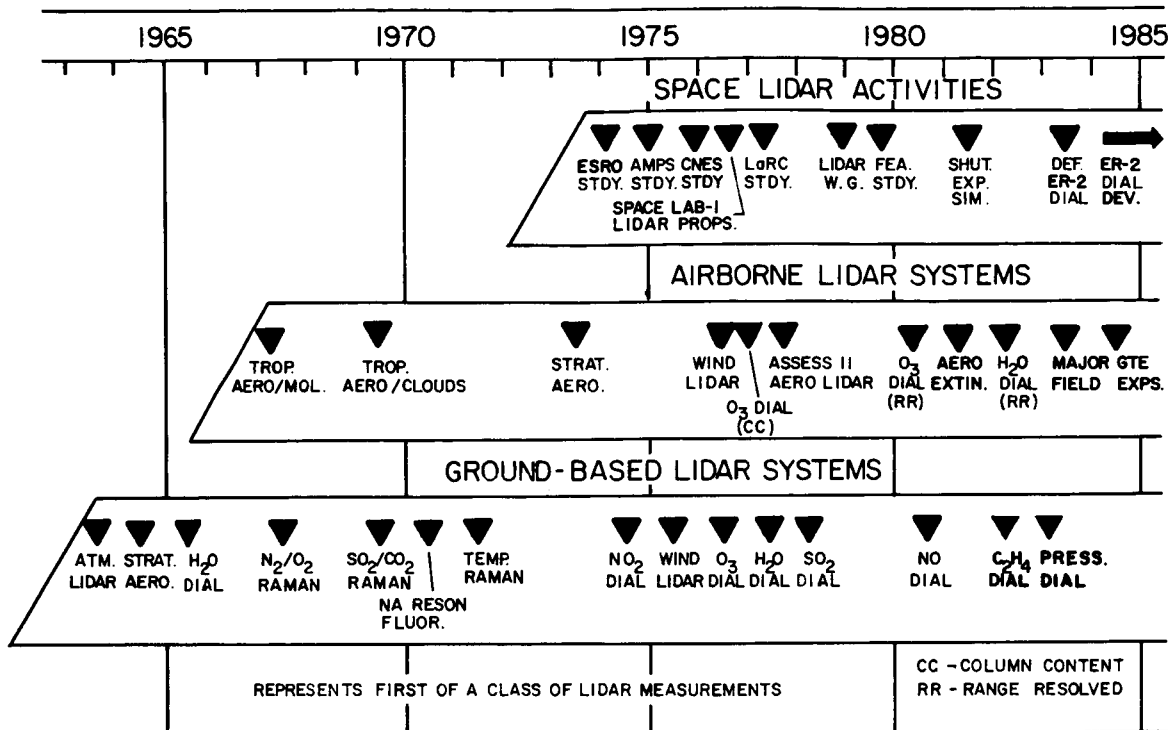


Figure 20.- History of lidar development with each milestone representing the first in a class of lidar measurements. In the case of ground-based and airborne lidar measurements the measurement parameter/species are given along with the lidar technique used. Various studies are cited in the activities supporting spaceborne lidar system development.

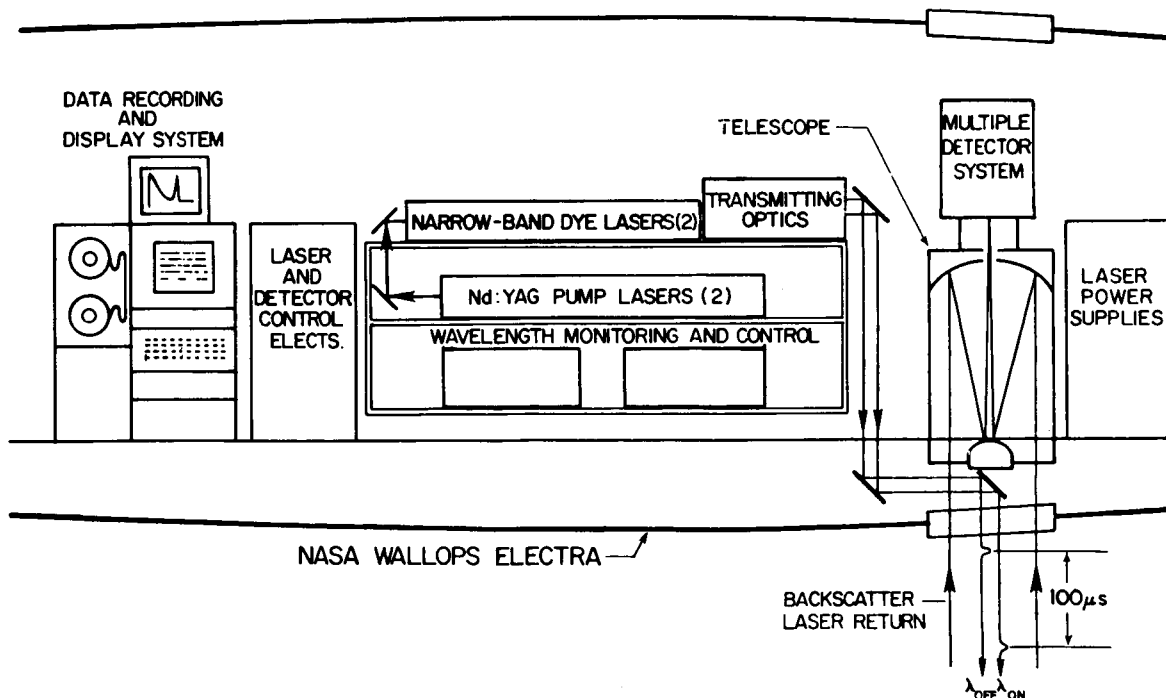


Figure 21.- NASA Langley Research Center airborne DIAL system schematic (Browell et al., 1983).

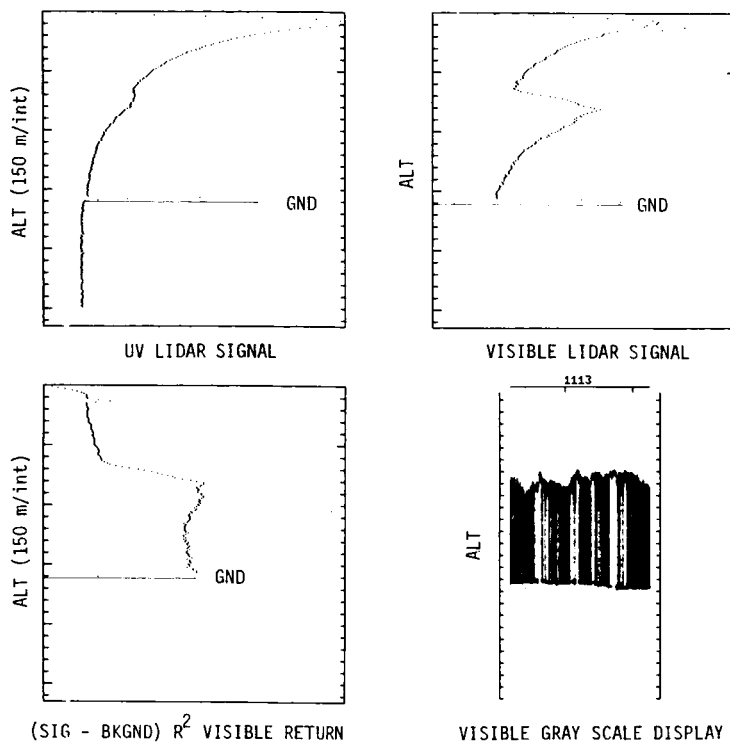


Figure 22.- Airborne DIAL UV and visible lidar returns with DIAL system operating in a nadir mode from an aircraft altitude of 3 km.

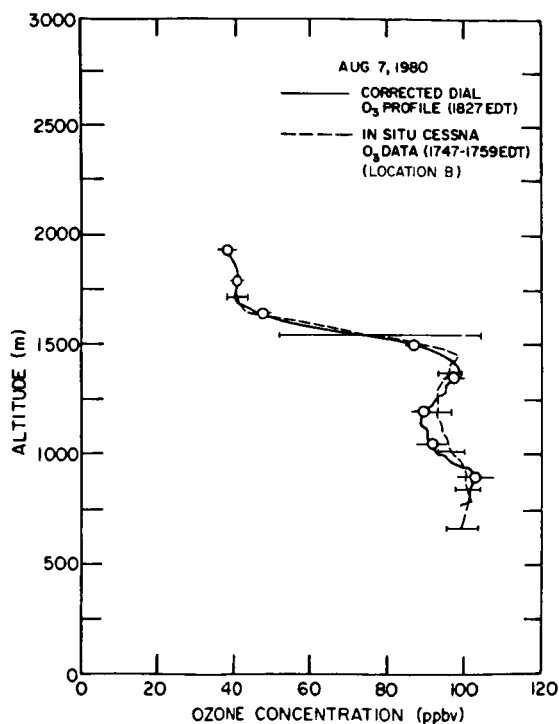


Figure 23.- Comparison of airborne DIAL and in situ measurements of O₃ profiles (Browell et al., 1985).

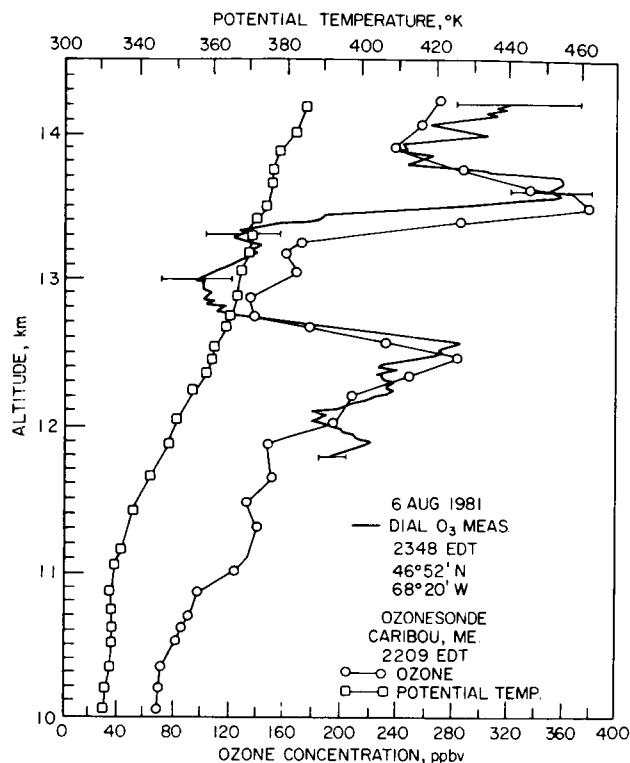


Figure 24.- Airborne DIAL and in situ O_3 measurements in the lower stratosphere (Browell, 1983).

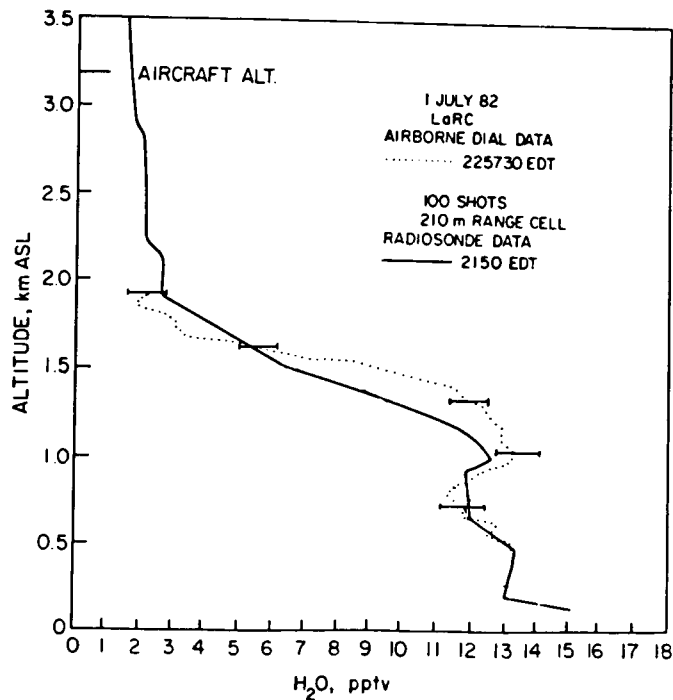


Figure 25.- Comparison of airborne DIAL and radiosonde measurements of H_2O profiles.

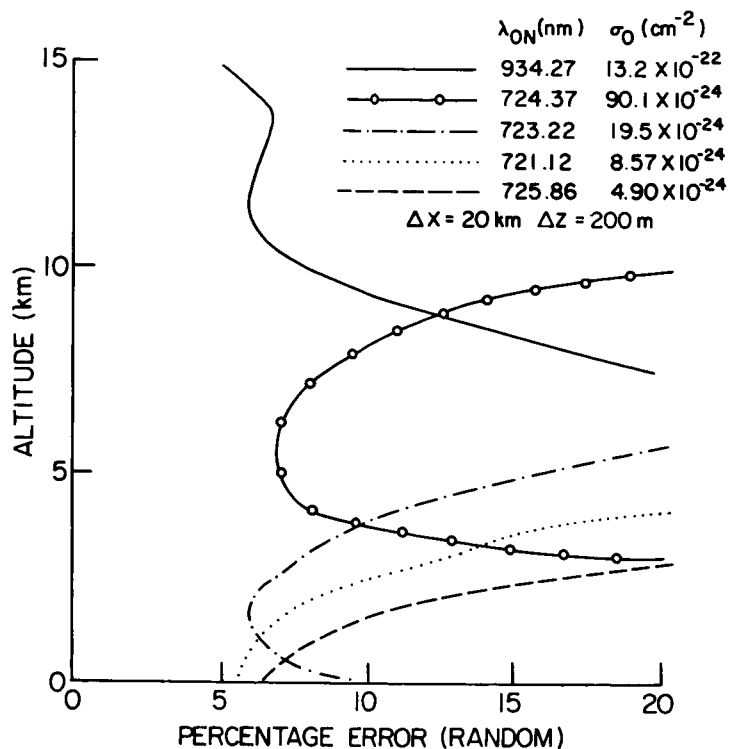


Figure 26.- Simulations of H₂O measurement uncertainties for the ER-2 DIAL system. An aircraft altitude of 16 km and a mid-latitude summer H₂O profile was assumed.

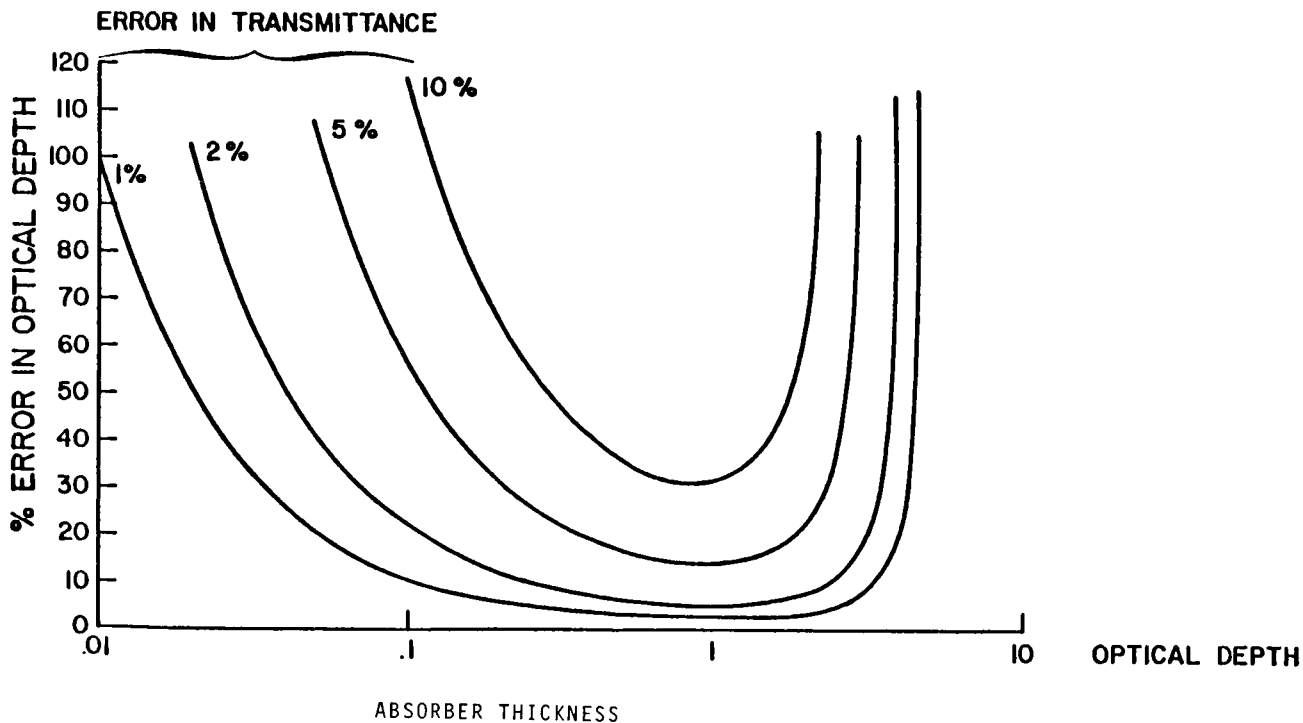


Figure 27.- Dependence of optical depth measurement error on absorber optical depth and transmittance measurement accuracy.

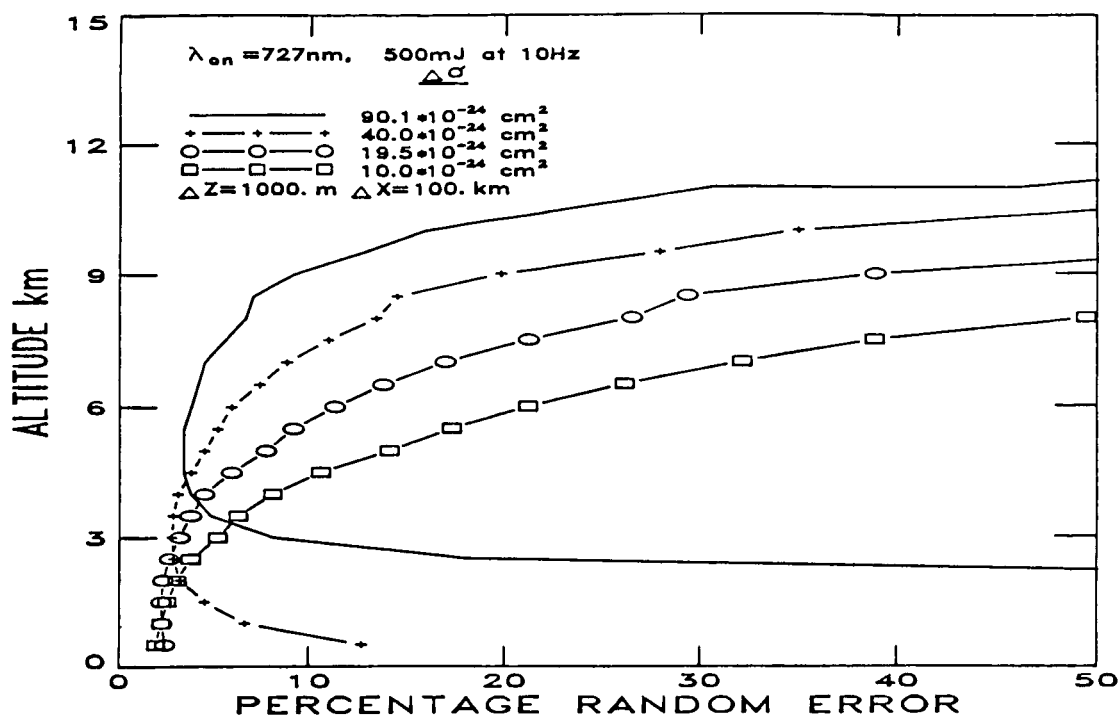


Figure 28.- Spaceborne DIAL H₂O profile measurement uncertainties. An orbital altitude of 800 km and a mid-latitude summer H₂O model profile were assumed.

APPENDIX A: TROPOSPHERIC CHEMISTRY: AN OVERVIEW

Joel S. Levine

OXYGEN SPECIES

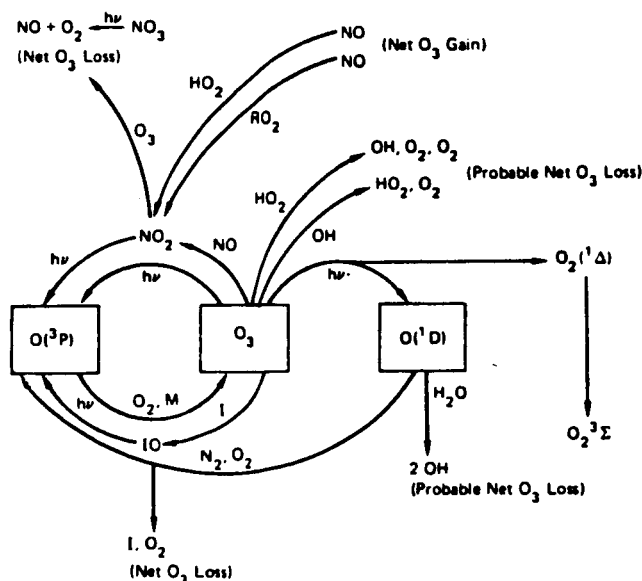
SPECIES	CONCENTRATION ¹	LIFETIME ²	COMMENTS ³
OZONE (O ₃)	10 - 100 PPBV	WEEKS TO MONTHS	C, TRANSPORT FROM STRATOSPHERE; GREENHOUSE SPECIES
ATOMIC OXYGEN (O)	10 ³ CM ⁻³	V.S.	C, LEADS TO O ₃ PRODUCTION
EXCITED ATOMIC OXYGEN (O(¹ D))	10 ⁻² CM ⁻³	V.S.	C, LEADS TO OH PRODUCTION

¹LEVINE, J.S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

²IBID, P. 44: V.S. = VERY SHORT (SECONDS OR LESS); V.L. = VERY LONG (DECADES OR LONGER)

³A = ANTHROPOGENIC; B = BIOGENIC; C = CHEMICAL OR PHOTOCHEMICAL

OXYGEN SPECIES CHEMISTRY



Photochemical production and destruction of O₃.
Key oxygen species are shown in solid boxes.

FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.

HYDROGEN SPECIES

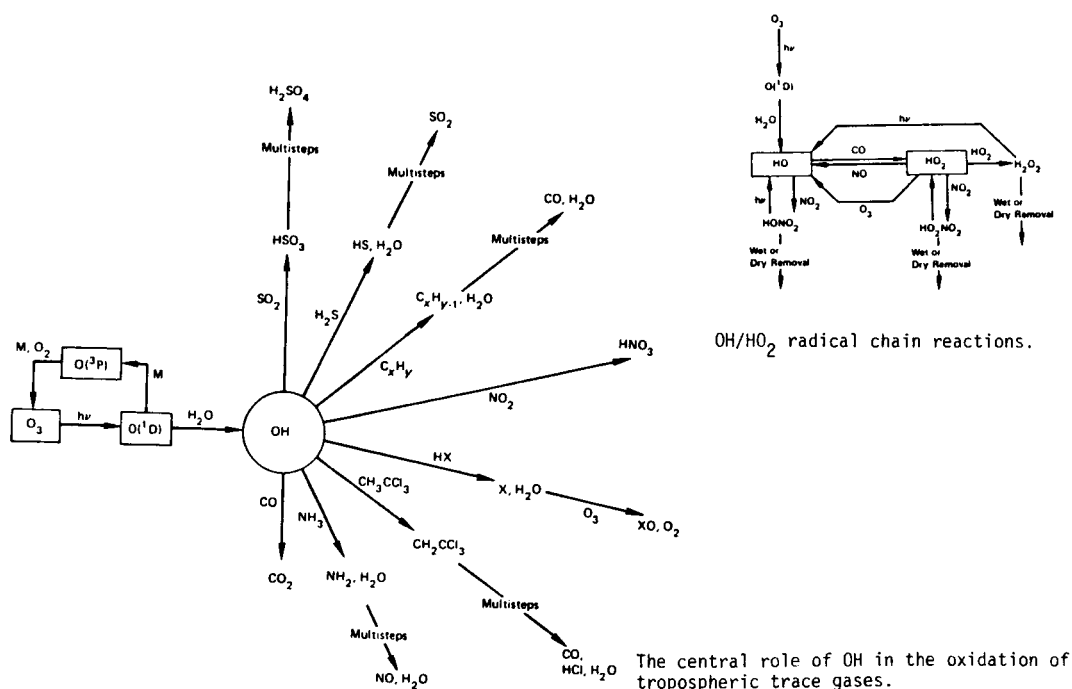
SPECIES	CONCENTRATION ¹	LIFETIME ²	COMMENTS ³
WATER VAPOR (H ₂ O)	<< 1% - 4%	HYDROLOGICAL CYCLE: VAPOR FORM: ~10 DAYS DROPLET FORM: ~HOURS	EVAPORATION/CONDENSATION; LEADS TO OH PRODUCTION; GREENHOUSE SPECIES
MOLECULAR HYDROGEN (H ₂)	0.5 PPMV	3.0×10^7 s (8.3×10^3 HR)	C, B
HYDROGEN PEROXIDE (H ₂ O ₂)	10^9 CM ⁻³	1.2×10^5 s (33 HR)	C; WATER SOLUBLE
HYDROPEROXYL RADICAL (HO ₂)	10^8 CM ⁻³	MINUTES	C
HYDROXYL RADICAL (OH)	10^6 CM ⁻³	V.S.	C; PRINCIPAL OXIDIZER IN TROPOSPHERE
ATOMIC HYDROGEN (H)	~ 1 CM ⁻³	V.S.	C

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HYDROGEN SPECIES CHEMISTRY



FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.

NITROGEN SPECIES

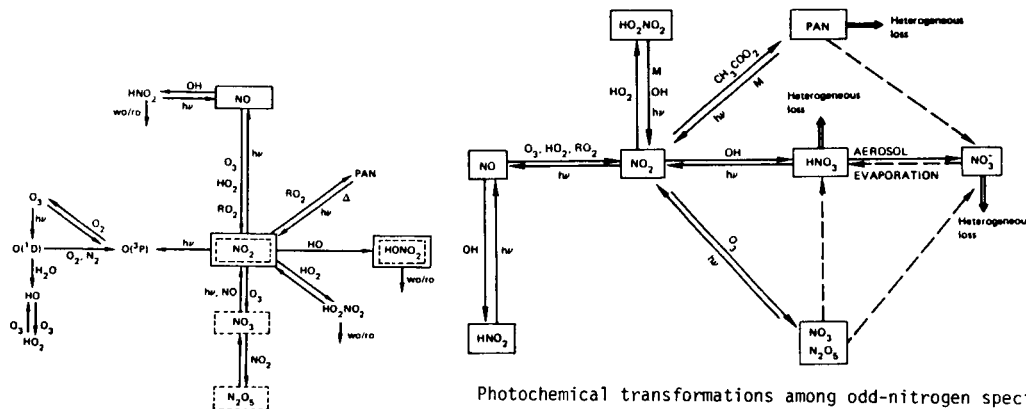
SPECIES	CONCENTRATION ¹	LIFETIME ²	COMMENTS ³
NITROUS OXIDE (N ₂ O)	330 PPBV	V.L.	B, A; GREENHOUSE SPECIES; GLOBAL INCREASE (~0.22/YR)
AMMONIA (NH ₃)	0.1 - 10 PPBV	1.3 x 10 ⁶ s (360 HR)	B, A; WATER SOLUBLE; MAJOR GASEOUS BASE; GREENHOUSE SPECIES; AEROSOL PRODUCTION
NITRIC ACID (HNO ₃)	50 - 1000 PPTV	1.5 x 10 ⁶ s (420 HR)	C; WATER SOLUBLE; ACID RAIN; AEROSOL PRODUCTION
NITROGEN DIOXIDE (NO ₂)	10 - 300 PPTV	1.3 x 10 ⁴ s (3.6 HR)	C
NITRIC OXIDE (NO)	5 - 100 PPTV	1.1 x 10 ² s (0.03 HR)	A, B, C, LIGHTNING
NITROGEN TRIOXIDE (NO ₃)	100 PPTV	-SECONDS	C
PEROXYACETYLNITRATE (CH ₃ CO ₃ NO ₂)	50 PPTV	DAYS TO YEARS	C
DINITROGEN PENTOXIDE (N ₂ O ₅)	1 PPTV	-SECONDS	C
PERNITRIC ACID (HO ₂ NO ₂)	0.5 PPTV	-DAYS	C; WATER SOLUBLE
NITROUS ACID (HNO ₂)	0.1 PPTV	-DAYS	C; WATER SOLUBLE

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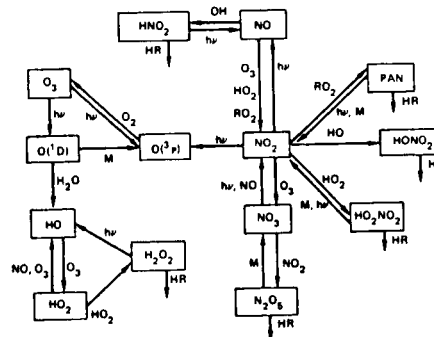
NITROGEN SPECIES CHEMISTRY



Photochemical transformations among odd-nitrogen species.

Major atmospheric reactions of N_xO_y species: Solid line boxes indicate major daytime nitrogen species; broken line boxes indicate significant nighttime nitrogen species. It is still uncertain whether PAN is a major species in the free troposphere. The notation wo/ro denotes washout/rainout process.

FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.



Major atmospheric reactions of H_xO_y, N_xO_y, and O₃, where M denotes N₂ and O₂, HR refers to heterogeneous removal, and hv indicates radiation required.

CARBON SPECIES

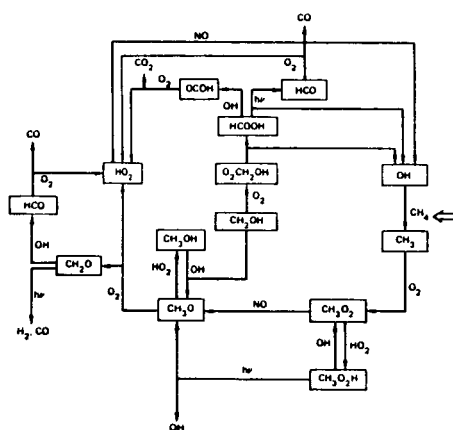
SPECIES	CONCENTRATION ¹	LIFETIME ²	COMMENTS ³
CARBON DIOXIDE (CO ₂)	0.034%	V.L.	A, B, VOLCANIC; GREENHOUSE SPECIES; GLOBAL INCREASE (~.2%/YR)
METHANE (CH ₄)	1.7 PPMV	2.5 x 10 ⁷ s (6.9 x 10 ³ HR)	B, A; GREENHOUSE SPECIES; GLOBAL INCREASE (~1.1%/YR)
CARBON MONOXIDE (CO)	70 - 200 PPBV (N.H.) 40 - 60 PPBV (S.H.)	7.1 x 10 ⁵ s (200 HR)	A, B, C; GLOBAL INCREASE (~1-2%/YR)
FORMALDEHYDE (H ₂ CO)	0.1 PPBV	~HOURS	C; WATER SOLUBLE
METHYHYDROPEROXIDE (CH ₃ OOH)	10 ¹¹ CM ⁻³	~DAYS	C; WATER SOLUBLE
METHYLPEROXYL RADICAL (CH ₃ O ₂)	10 ⁸ CM ⁻³	V.S.	C
METHYL RADICAL (CH ₃)	10 ⁻¹ CM ⁻³	V.S.	C

¹LEVINE, J.S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

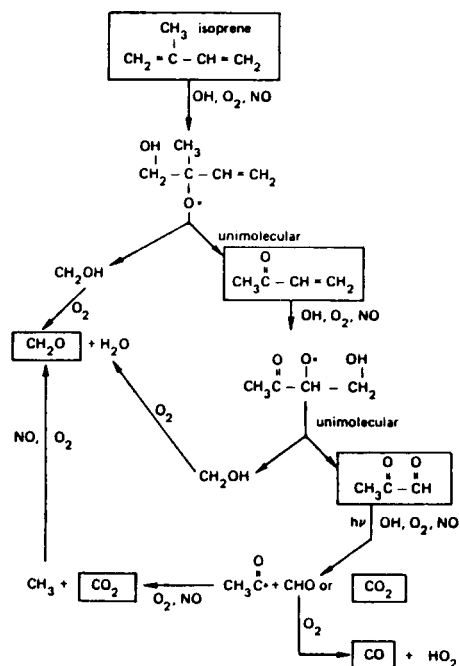
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CARBON SPECIES CHEMISTRY



A possible tropospheric degradation scheme for CH₃O₂ radicals, formed from CH₄. The current lack of understanding of this chemistry defines one of the major uncertainties in the understanding of fast H_xO_y photochemistry.



A possible reaction scheme for isoprene oxidation in the presence of NO_x.

FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.

SULFUR SPECIES

SPECIES	CONCENTRATION ¹	LIFETIME ²	COMMENTS ³
CARBONYL SULFIDE (COS)	0.5 PPBV	$\geq 2.2 \times 10^7$ s ($\geq 1 \times 10^3$ HR)	VOLCANIC, A
DIMETHYL SULFIDE ((CH ₃) ₂ S)	0.4 PPBV	-HOURS	B
HYDROGEN SULFIDE (H ₂ S)	0.2 PPBV	3.8×10^4 s (11 HR)	B, A
SULFUR DIOXIDE (SO ₂)	0.2 PPBV	8.0×10^4 s (22 HR)	VOLCANIC, A, C; AEROSOL PRODUCTION
DIMETHYL DISULFIDE ((CH ₃) ₂ S ₂)	100 PPTV	-HOURS	B
CARBON SULFIDE (CS ₂)	50 PPTV	-DAYS	VOLCANIC, A
SULFURIC ACID (H ₂ SO ₄)	20 PPTV	-DAYS	C; WATER SOLUBLE; ACID RAIN
SULFUROUS ACID (H ₂ SO ₃)	20 PPTV	-DAYS	C
SULFOXYL RADICAL (SO)	10^3 CM ⁻³	V.S.	C
THIOHYDROXYL RADICAL (HS)	1 CM ⁻³	V.S.	C
SULFUR TRIOXIDE (SO ₃)	10^{-2} CM ⁻³	V.S.	C

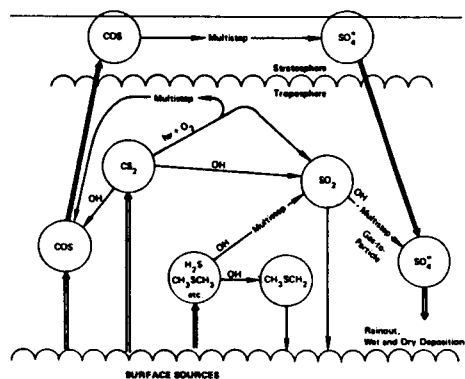
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SULFUR SPECIES CHEMISTRY

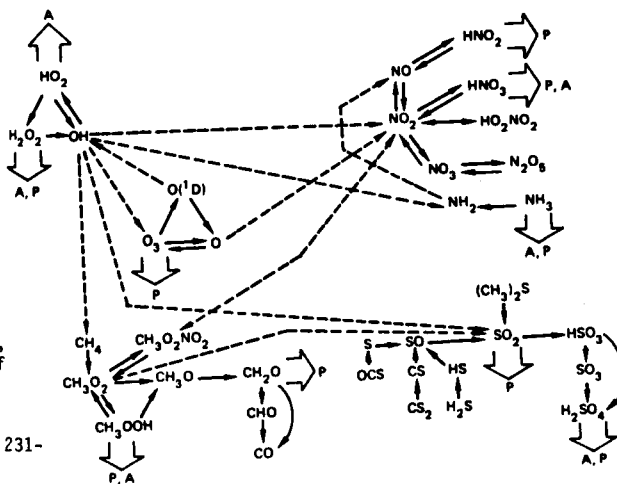


A tentative scheme for the oxidation and removal of atmospheric sulfur species.

FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.

Gas-phase constituents and major reaction pathways (solid lines). Interactions between chemical families are indicated by dashed lines. Heavy (double) arrows show key heterogeneous pathways involving aerosols (A) and precipitation (P). Turco et al., 1982.*

*Turco, R. P., O. B. Toon, R. C. Whitten, R. G. Keese and P. Hamill, Importance of Heterogeneous Processes to Tropospheric Chemistry, Geophysical Monograph Series, vol. 26, D. R. Schryer, editor, American Geophysical Union, Washington, D.C., pp. 231-240, 1982.



HALOGEN SPECIES

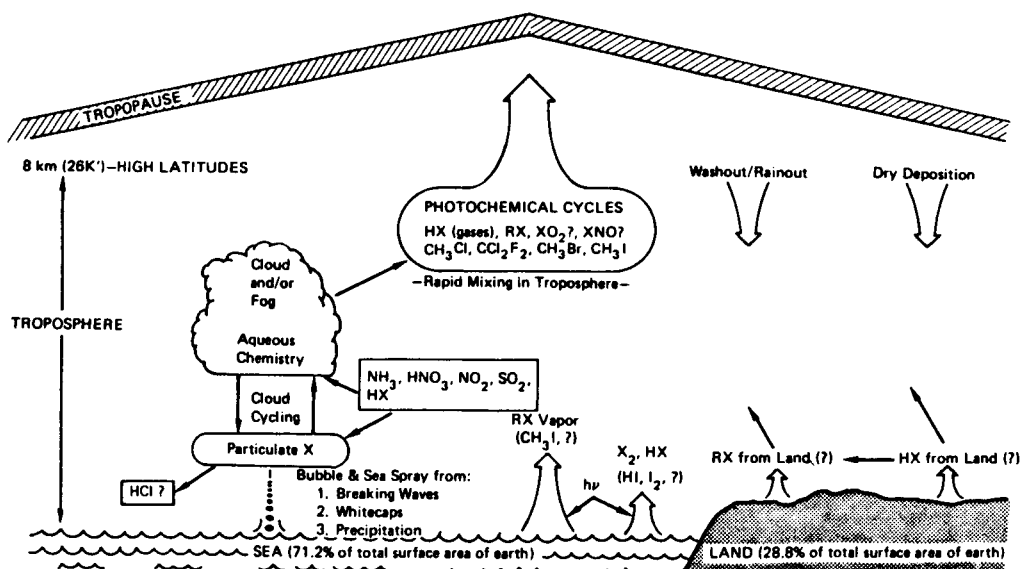
SPECIES	CONCENTRATION ¹	LIFETIME ²	COMMENTS ³
HYDROGEN CHLORIDE (HCl)	1 PPBV	3.0×10^5 s (83 HR)	SEA SALT, VOLCANIC; WATER SOLUBLE
METHYL CHLORIDE (CH ₃ Cl)	0.5 PPBV	4.9×10^6 s (1.3×10^3 HR)	B, A
METHYL BROMIDE (CH ₃ Br)	10 PPTV	~MONTHS	B, A
METHYL IODIDE (CH ₃ I)	1 PPTV	~MONTHS	B, A

¹LEVINE, J.-S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

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HALOGEN SPECIES CHEMISTRY

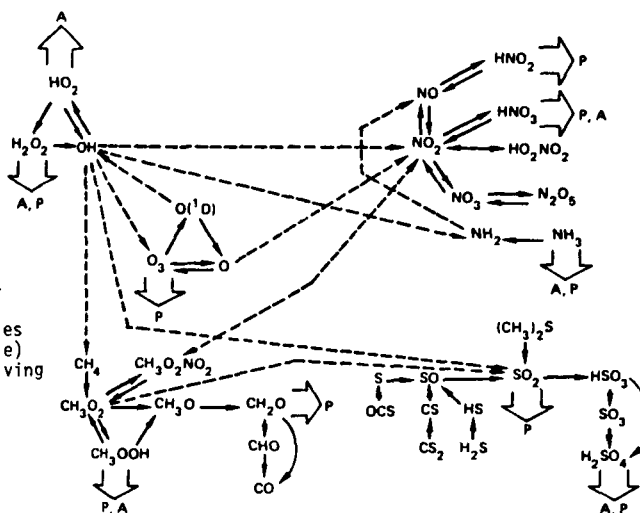


Schematic diagram to show processes and to exemplify key species in global tropospheric halogen cycles. X denotes F, Cl, Br, or I.

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Aerosols as an end product of atmospheric reactions. Major reaction pathways for gas-phase constituents are depicted by solid lines. Interactions between chemical families are indicated by dashed lines. Heavy (double) arrows show key heterogeneous pathways involving aerosols (A) and precipitation (P). Turco et al., 1982.*

Turco, R. P., O. B. Toon, R. C. Whitten, R. G. Keese and P. Hamill, Importance of Heterogeneous Processes to Tropospheric Chemistry, Geophysical Monograph Series, vol. 26, D. R. Schryer, editor, American Geophysical Union, Washington, D.C., pp. 231-240, 1982.



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